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Areal Distribution of ^{60}Co, ^{137}Cs, and ^{90}Sr in Streambed Gravels of White Oak Creek Watershed Oak Ridge, Tennessee

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ENVIRONMENTAL SCIENCES DIVISION
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AREAL DISTRIBUTION OF ^{60}Co, ^{137}Cs, AND ^{90}Sr IN STREAMBED GRAVELS OF WHITE OAK CREEK WATERSHED OAK RIDGE, TENNESSEE

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Publication No. 1488

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ABSTRACT

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A comprehensive survey was performed of the concentrations of ^{90}Sr, ^{60}Co, and ^{137}Cs in streambed gravels from contaminated drainages in White Oak Creek Watershed. Methods to interpret these concentrations in terms of the relative contributions of various sources to the total discharge from the watershed were developed. Principal sources of ^{90}Sr, as a percent of the total discharge at the time of sampling, were: direct ORNL plant effluents (50%), leaching from solid waste disposal area (SWDA) 4 (30%), and leaching from SWDA 5 (10%). Minor sources included SWDA 3, the Molten Salt Reactor Facility, and intermediate-level liquid waste pit 1 with each representing 4% or less of the total basin discharge. The cooling water effluent from the High-Flux Isotope Reactor was the dominant source of ^{60}Co contamination in the watershed. ORNL plant effluents accounted for almost all the ^{137}Cs discharge from White Oak Creek basin. Point sources of contamination led to constant downstream radionuclide concentrations until significant dilution by other tributaries occurred. The extent of present contamination throughout the watershed was delineated such that any future activities giving rise to additional contamination can be identified.

Distribution coefficients, K_d's, between streambed gravels and streamwater were determined for ^{85}Sr, ^{60}Co, and ^{137}Cs: 50, 560,

and 8460 ml/g, respectively (mean of 24 samples). An abridged radiochemical fractionation for ^{90}Sr was developed involving a single carbonate and nitrate precipitation of Sr carrier; it was found to be as accurate and precise for these samples as the standard ^{90}Sr method above levels of 2 dpm/g.

TABLE OF CONTENTS

LIST OF FIGURES

INTRODUCTION

White Oak Creek Watershed contains the numerous facilities and activities of the Oak Ridge National Laboratory (ORNL) which give rise to discharges of radioisotopes into the public environment. The watershed discharges into the Clinch River just below White Oak Dam where radionuclide concentrations are continuously monitored. At this discharge point, all radioisotopes, except ^{90}Sr, are present at concentrations in orders of magnitude generally below currently recommended maximum permissible concentrations (MPC's). The MPC's for ^{60}Co, ^{137}Cs, and ^{90}Sr for the unrestricted use of water are 111, 44, and 0.67 dpm/ml, respectively (Brodsky 1969). However, ^{90}Sr concentration at White Oak Dam has been and continues to be near, and sometimes in excess of, the MPC at this point. It should be pointed out that the ^{90}Sr concentration is diluted several hundredfold by the Clinch River thus bringing its concentration well below MPC in the river. Nevertheless, it is the general goal of the radioactive waste management program of the ORNL to develop techniques to reduce the discharge of ^{90}Sr and other radionuclides.

Before corrective action can be taken, it is desirable to know all sources of ^{90}Sr within the watershed and their contributions to the total discharge. Through the routine monitoring of water flow and ^{90}Sr concentrations at permanent monitoring stations at various points in the watershed (Fig. 1), most of the major sources of ^{90}Sr have been identified within particular areas. These known sources include ORNL plant effluents, SWDA 4, and SWDA 5 (Fig. 1). Until

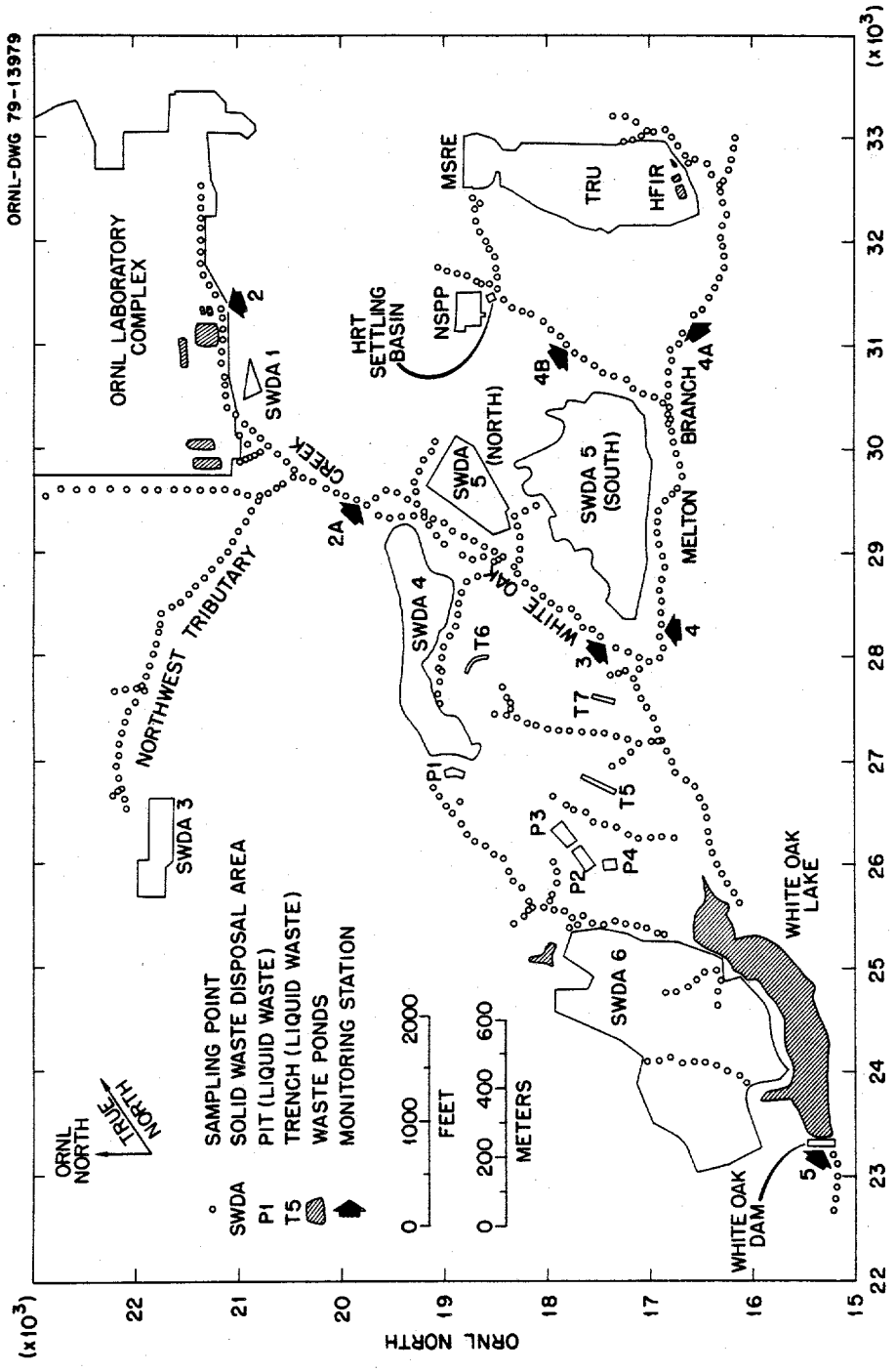

Fig. 1. White Oak Creek watershed indicating solid waste disposal areas, liquid waste pits and trenches, permanent monitoring stations, and sampling points used in this study.

recently, most of the ^{90}Sr discharge originated from direct ORNL plant effluents (Stueber et al. 1978). Reductions in these plant effluents should increase the relative contributions from the other, presumably, more diffuse sources such as the SWDA's.

The watershed contains four large solid waste disposal areas (Fig. 1): SWDA 3, a 2.8-ha site operated from 1946 to 1951; SWDA 4, a 9.3-ha site operated from 1951 to 1959; SWDA 5, a 13.3-ha site operated from 1958 to 1973; and SWDA 6, a 28.3-ha site in use from 1973 to the present. In addition, there are seven seepage pits and trenches, last used in the mid-1960's, for the disposal of intermediate-level liquid waste. Other potential sources also exist in the watershed outside the main plant complex including the High-Flux Isotope Reactor (HFIR), the Homogeneous Reactor Test (HRT) settling basin near what is now known as the Nuclear Safety Pilot Plant (NSPP) building, the Molten-Salt Reactor Experiment (MSRE) building, and the Transuranium Processing Plant and Thorium-Uranium Recycle Facility (TRU). In addition, White Oak Lake, created by White Oak Dam, has been functioning for over thirty years as a solids-settling reservoir; it may be presently functioning as a source of radionuclide discharge.

Although the contribution to the total ^{90}Sr discharge from some of these general sources is known, the precise location within these larger areas is generally not known. In addition, minor sources of ^{90}Sr were recently found in several smaller areas of the watershed (Spalding and Cerling 1979); the contribution of these areas to the total discharge is not known but is presumed to be small. Our recent study (Spalding and Cerling 1979) on the mechanisms of radionuclide

adsorption by streambed sediments of White Oak Creek pointed out the advantages of using the coarse sand to fine gravel fraction of these sediments to locate sources of contamination. These advantages included the much higher concentrations of radionuclides in the gravels than the associated streamwater and the relative (to water and finer particle sizes) stability of the gravels to downstream movement. That study also included a preliminary survey of several drainages within the watershed and pointed out the need for a much more comprehensive survey of the entire watershed. The present study reports the concentrations of ^{90}Sr, ^{60}Co, and ^{137}Cs in all contaminated tributaries sampled at approximately 35-m intervals. It also represents the first comprehensive survey of the entire watershed made over a short period of time. It should function as a future reference to compare the effectiveness of any corrective measures applied to reduce the discharge from any of the various sources. Of equal importance, this survey points out the extent of present contamination: any future activities of the laboratory which contaminate other areas of the watershed may be delineated from the previously contaminated areas noted in this survey.

We also present a method to interpret these radionuclide concentrations of gravels in terms of the relative contributions of various drainages within the watershed to the total discharge of each radionuclide. White Oak Creek watershed can be divided into smaller drainage areas which differentiate various known and potential sources of contamination (Fig. 2). The drainage divides delineated in Fig. 2 were chosen to group areas which drain into contaminated reaches of

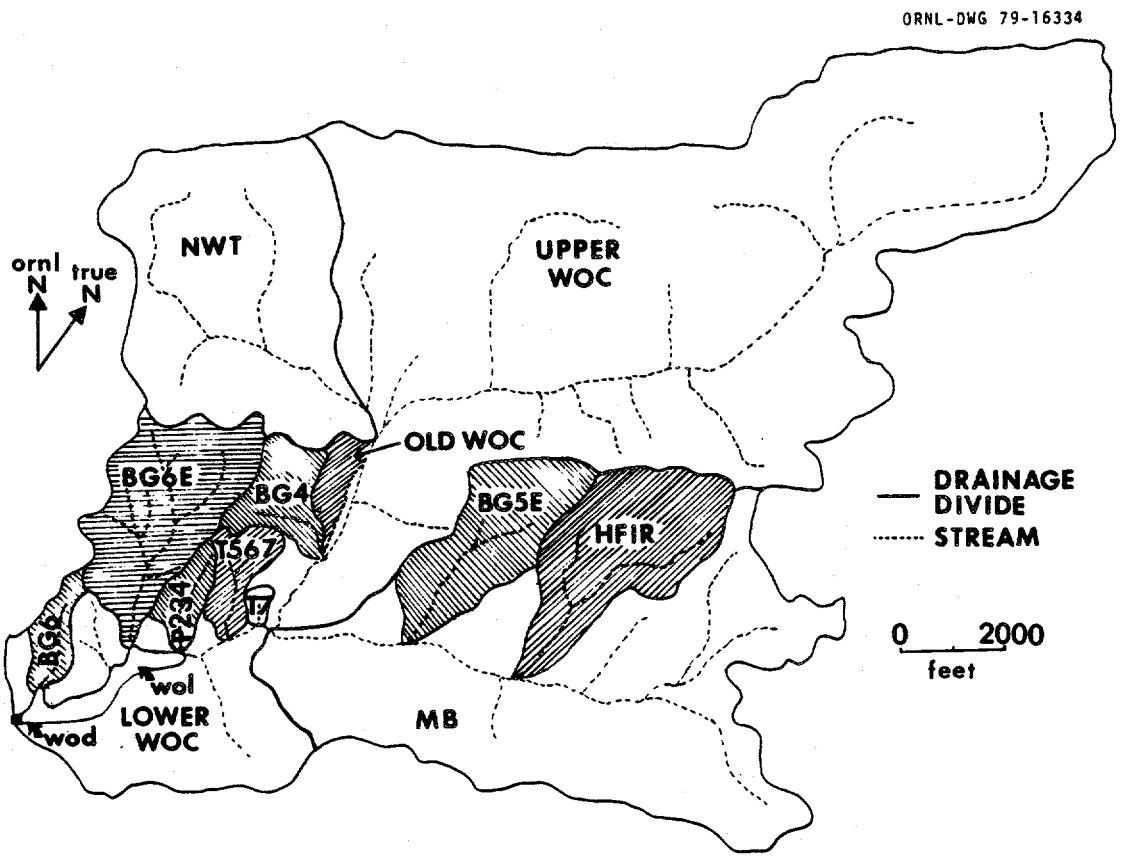

Fig. 2. Subdrainages of interest in radionuclide contamination in White Oak Creek basin: NWT-drainage of SWDA 3 through the Northwest Tributary; WOC-White Oak Creek drainage, upper and lower; HFIR-drainage through the high-flux isotope reactor complex; BG5E-drainage east of SWDA5; MB-drainage through Melton Branch other than HFIR and BG5E; Wol-White Oak Lake; Wod-White Oak dam; BG6-drainage from central SWDA6; BG6E-drainage from east of SWDA6; P234-drainage from seepage pits 2, 3, and 4; T567-drainage from waste trenches 5, 6, and 7; T7 drainage east of trench 7; BG4-drainage from SWDA4; OLD WOC - Drainage from contaminated floodplain through old channel of White Oak Creek.

creeks. Assuming that the water discharge from a given reach is proportional to the area drained by that reach, the radionuclide discharge from different drainages can be compared at the time of sampling. The mean streamwater concentration of a radionuclide can be estimated from laboratory-measured K_d's (equilibrium distribution coefficients of radionuclide between streamwater and gravel) by dividing into the mean gravel concentrations near the mouth of these drainages. The products of these mean streamwater concentrations and the areas drained provide relative values to compare and rank the discharges from these various areas. The value of the K_d used in these calculations will depend on the mineralogical composition of the gravel. Gravels in the upper half of the basin, including the Northwest tributary and upper White Oak Creek (Fig. 2), are predominantly composed of chert and limestone fragments. The upper part of the watershed in Bethel Valley is underlain by Chickamauga limestone and Knox dolomite bedrock (Webster 1979). The Bethel Valley drainage flows into the southern half of the watershed in Melton Valley which is underlain by Conasauga shale bedrock. Therefore, increasing amounts of shale compose the gravels of White Oak Creek as it flows from Bethel into Melton Valley and on into White Oak Lake. The effects of these changes in mineralogy were determined by measuring K_d's for gravels for each drainage and radionuclide. Values of K_d, representative of the type of gravel, were then used to calculate the mean streamwater concentrations in each drainage.

Although ^{60}Co and ^{137}Cs discharges from the watershed are far below the MPC's (111 and 44 dpm/ml, respectively, compared to that of

^{90}Sr: 0.67 dpm/ml), their distributions in the watershed were also determined since their presence indicates where laboratory activities introduce contamination into the watershed. In addition, both radionuclides are of interest since their discharge could also be reduced if the major sources were known. Both ^{60}Co and ^{137}Cs concentrations in gravel are easy to measure via direct gamma-ray spectrometry. On the other hand, ^{90}Sr analyses must be performed on gravel extracts and are both time-consuming and expensive due to the involved radiochemical fractionation to separate out other fission and activation products (American Public Health Service 1975). In the present study, we have developed a considerably abridged radiochemical fractionation for the determination of ^{90}Sr and have demonstrated its applicability to our samples. This abridged procedure was then employed for the ^{90}Sr analyses of most of our gravel samples; it represented a considerable savings in time with no compromise in either accuracy or precision for samples with ^{90}Sr levels above atmospheric fallout background.

METHODS

Samples of streambed gravel were collected from all major streams and their significant tributaries in White Oak Creek basin during October and November 1978. An additional fifty samples were collected in February 1979 to more precisely locate sources of contamination revealed in the initial suite of 362 samples. Gravels were collected from the upper 10 cm of streambed sediment and were wet sieved (6 to 20 mesh; 3.35 to 0.85 mm) directly in their streamwater. In the few

samples from dry stream beds of seasonally intermitent streams, the gravels were sieved in the nearest pool or stream. Samples were collected approximately every 35 m and locations were marked with flags placed in the nearest bank. Sample locations relative to each other were established with a compass bearing and a distance measured with an optical rangefinder (Leitz). Significant landmarks (roads, permanent monitoring stations, stream junctions, etc.) were also included to establish absolute locations by reference to a recently prepared topographic map of the watershed (Accu-Air Survey 1978). Each sampling location was estimated to be within 3 m of the map grid coordinates listed in the Appendix. These sampling points are depicted in Fig. 1.

Each gravel sample was dried at 70°C in a forced-air oven for 18 h. A ten-gram aliquot was placed in a 30-ml glass scintillation vial and counted directly for ^{60}Co and ^{137}Cs in a 3 x 3 in. NaI(Tl) well-type detector using a Packard Model 5320 Auto Gamma Scintillation Spectrometer equipped with a Packard Model 9012 Multichannel Analyzer. Standards were prepared by adding 100 µliters of certified standard reference solutions (Amersham Radiochemicals) to 10 g of uncontaminated gravel. Baseline corrections for a given photopeak were calculated by substracting the average counts per channel for the twenty channels on either side of the photopeak multiplied by the number of channels in the photopeak. The validity of this baseline correction was confirmed for mixed ^{60}Co-^{137}Cs standards whose activities agreed with otherwise identical single radioisotope standards. Detection limits were estimated to be 0.5 dpm/g of gravel for both isotopes; this estimate was based on the counting time (typically 20 min), the counting

efficiencies (14.7 and 9.5% for ^{137}Cs and ^{60}Co, respectively), and baseline correction (which varied for each sample depending on the relative amounts of each isotope).

A second 10-g subsample was used for the analyses of ^{90}Sr, Fe, and Mn. This gravel was extracted three times with 30 ml of 2% hydroxylamine hydrochloride in 0.3 M ammonium citrate adjusted to pH 7.0 with NH_4OH (Whitney 1975) at 90°C. The supernatants from each extraction, after centrifugation at 1,100 RCF for 10 min, were combined for each sample and adjusted to 100 ml with the above solution. Iron and manganese were determined colorimetrically on these extracts (Olson 1965; Adams 1965). An abbreviation of the standard method for the determination of radiostrontium in water (American Public Health Service 1975) was employed for the ^{90}Sr determination in these extracts:

(1) Add 2 ml of $Sr(NO_3)_2$ carrier to 25 ml of extract and heat in 95°C waterbath for 30 min.

(2) Add 10 ml 6 N NaOH and 5 ml 2 N Na_2CO_3 and heat for 30 min. Add a second 5 ml 2 N Na_2CO_3 and heat an additional 30 min.

(3) Centrifuge at 300 RCF for 10 min and discard supernatant. Dissolve pellet by adding 4 ml conc. HNO_3, heat 5 min until dissolved, and cool in ice bath.

(4) Add 20 ml fuming HNO_3, cool 10 min in ice bath, centrifuge, discard supernatant, and drain excess HNO_3.

(5) Suspend pellet with 20 ml acetone, centrifuge, discard supernatant, and allow excess acetone to evaporate.

(6) Add 1.5 ml water, dissolve pellet, heat 2 min, and transfer to a one-inch stainless steel planchet, previously tared.

(7) Dry under infrared lamp, cool, weigh, and count after three weeks. A Beckman Wide-Beta II gas-flow proportional counter was used to determine ^{90}Sr activities on these planchets. Counting efficiency was determined from a self-absorption curve of known ^{90}Sr activity with increasing total solids per planchet. An average yield for this procedure was calculated based on recovery of known additions of ^{90}Sr to 41 randomly selected samples. In addition, 31 samples (selected to cover the range of ^{90}Sr concentrations encountered) were analyzed via the standard method (American Public Health Service 1975) to compare with the results of this abridged method. Known additions of ^{137}Cs and ^{60}Co were also prepared in water free of ^{90}Sr to determine their degree of radiochemical separation from ^{90}Sr in this abridged procedure; since both ^{137}Cs and ^{60}Co exhibit beta activity, they pose a potential interference with the ^{90}Sr determination unless they are removed.

The equilibrium distribution coefficients, K_d's, for ^{85}Sr, ^{60}Co, and ^{137}Cs between various streambed gravels and streamwater were also determined. Five grams of each gravel were placed in a 30-ml polypropylene "Oak Ridge" centrifuge tube with 25 ml of streamwater freshly collected at White Oak Dam on August 30, 1979. This water had a pH = 6.6 and an electrical conductivity = 280 μmhos/cm and an EDTA hardness of 123 mg $CaCO_3$/liter. To each tube was added one ml of stream water (^{85}Sr) or tap water (^{60}Co and ^{137}Cs) containing the carrier-free radioisotope at an activity of 10^6 dpm/ml or greater.

The tubes were shaken for either 24 hr (^{85}Sr) or 120 hr (^{60}Co and ^{137}Cs), centrifuged at 3,500 RCF for 10 min, and a 5-ml aliquot removed for activity determination. Gamma activity of this aliquot was determined using the counting procedure described previously. Blanks, without gravel but with either stream water or tap water, were run concurrently to determine the total activity without adsorption to the gravel. The K_d as then calculated:

$$K_d = \frac{\text{(fraction adsorbed)}}{1 - \text{(fraction adsorbed)}} \times \frac{\text{Volume (26 ml)}}{\text{Weight (5 g)}}$$

Gravel samples were selected for these K_d determinations to represent the different drainages and locations delineated in Fig. 2.

RESULTS

Abridged Method for ^{90}Sr Determination

Figure 3 shows the relation between the ^{90}Sr concentrations by the abridged and standard methods. Since the slope of the regression was essentially one and the correlation coefficient equal to 0.9996, the abridged method was a valid procedure to determine ^{90}Sr activity in these streambed sediment extracts. Known additions of ^{60}Co and ^{137}Cs indicated that only 0.05 and 0.009% of their activities, respectively, were carried on the final $Sr(NO_3)_2$ precipitate of the abridged procedure. Such yields would lead to undetectable interferences in the ^{90}Sr determination by ^{60}Co and ^{137}Cs, considering the levels of these radionuclides in the streambed gravels (appendix). The average yield of ^{90}Sr for the abridged procedure was

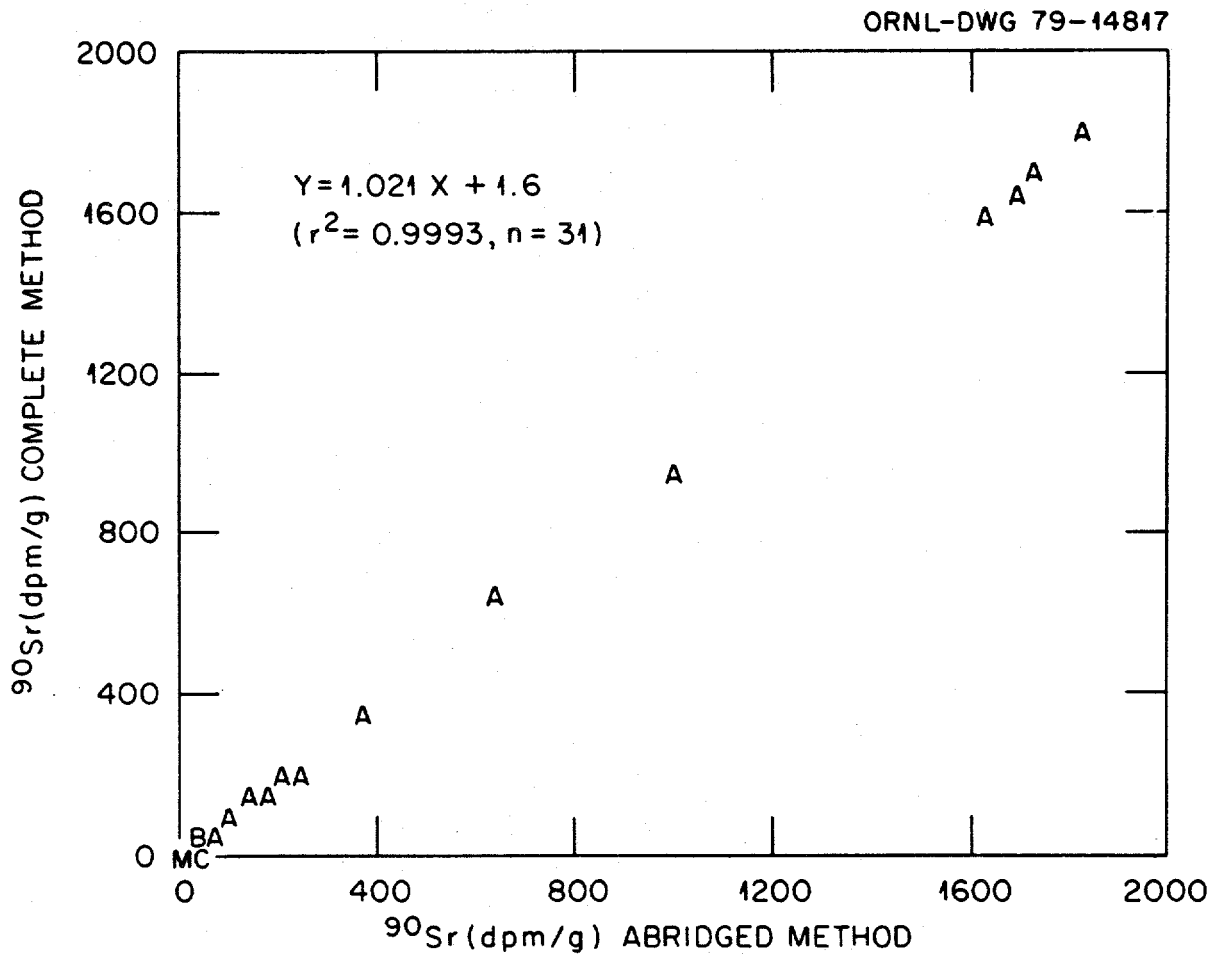

Fig. 3. Concentrations of ^{90}Sr in stream-bed gravel extracts determined by an abridged and standard radiochemical method.

86.5% ± 2.5% (1σ); the standard method typically yielded between 60 and 77% of the ^{90}Sr (based on the weighed recovery of Sr carrier). The major advantage of the abridged procedure was its savings in time and effort, i.e., three precipitations and one container transfer versus twelve precipitations and six container transfers in the standard method. This was achieved without sacrificing either yield (sensitivity) or accuracy. Such an abridged procedure worked well with these samples from White OaK Creek watershed since many samples contained levels of ^{90}Sr which were orders of magnitude greater than those produced by atmospheric fallout. The procedure would definitely not be applicable to fallout ^{90}Sr determinations where interferences from other radionuclides would not be adequately removed.

Distribution Coefficients for ^{85}Sr, ^{60}Co, and ^{137}Cs

The K_d's for ^{85}Sr, ^{60}Co, and ^{137}Cs between stream water and gravels are listed in Table 1. The average K_d's for ^{85}Sr, ^{60}Co, and ^{137}Cs were 50.3, 564, and 8460 ml/g, respectively. This illustrates the relative adsorption of these radionuclides by White Oak Creek sediments. In our previous study (Spalding and Cerling 1979) we quoted typical K_d values for Conasauga shale for ^{90}Sr ^{60}Co, and ^{137}Cs of 120, 70,000, and and 100,000 ml/g. The quoted ^{90}Sr K_d was based on our observations in that study, whereas the ^{60}Co and ^{137}Cs K_ds were taken from observations using distilled water and Clinch River sediment (Morton 1961) since the mineralogy of this sediment was very similar to White Oak Creek sediment. The K_d values in Table 1 were performed in stream water and hence these K_ds are

Table 1. Equilibrium distribution coefficients (K_d's) for ^{85}Sr, ^{60}Co, and ^{137}Cs between streambed gravels and stream water

Sample[a]	Watershed[b] location	K_d (ml/g) ^{85}Sr	^{60}Co	^{137}Cs
1	Upper WOC	13.4	227	3,130
11	Upper WOC	20.1	599	5,770
21	Upper WOC	16.5	379	3,210
31	Upper WOC	19.0	726	6,340
41	Upper WOC	17.2	308	2,090
51	Lower WOC	40.5	607	12,400
61	Lower WOC	34.5	448	8,120
71	Lower WOC	49.2	717	13,000
81	Lower WOC	54.8	827	11,600
91	Lower WOC	58.4	612	8,520
100	BG4	98.4	476	17,300
150	MB	61.5	851	14,400
175	HFIR	90.9	1,160	4,480
205	BG5E	76.5	843	23,600
240	BG6E	107.8	323	10,200
250	OLD WOC	24.9	363	4,680
260	NWT	22.6	303	3,850
270	NWT	38.4	475	8,000
280	NWT	31.8	320	3,910
290	NWT	27.3	448	6,260
305	P234	111.1	393	6,340
330	T567	78.0	890	11,800
335	Below wod	25.5	351	5,300
350	BG6	89.9	891	8,810
Average ± 1 σ		50.3 ± 31.7	564 ± 247	8,460 ± 5,130

[a]See Appendix for sample location and description.

[b]See Fig. 2 for description of watershed location code.

much lower than the quoted values for distilled water. Nevertheless, the values in Table 1 demonstrate the order of magnitude difference in K_d from ^{137}Cs to ^{60}Co and from ^{60}Co to ^{90}Sr. These average K_d's were used to calculate an average water concentration in the watershed simply by dividing them into the average sediment concentrations in the whole watershed, i.e., 70, 435, and 973 dpm/g for ^{90}Sr, ^{60}Co, and ^{137}Cs, respectively. This calculation yielded water concentrations of 1.4, 0.77, and 0.12 dpm/ml (212, 0.7, and 0.3% of MPC), respectively. Such a calculation serves only to put into perspective the relative importance of these three radionuclides in the radiocontamination of the watershed.

The utility of these K_d's can be further illustrated by calculating the average water concentration of ^{90}Sr entering White Oak Lake and, presumedly, discharging at White Oak dam. The mean ^{90}Sr concentration in the ten gravel samples immediately upstream from White Oak Lake was 32.5 dpm/g (samples 84 to 93, Appendix). The average K_d of the five samples (51, 61, 71, 81, and 91, Table 1) in lower White Oak Creek was 47.5 ml/g. The predicted stream-water concentration of ^{90}Sr would then be 0.68 dpm/ml or 103% of MPC. Notably, the average annual concentrations of ^{90}Sr at White Oak dam from 1974 to 1977 has ranged from 80 to 195% of MPC (Stueber et al. 1978). This agreement between calculated average concentration and the monitored concentrations at White Oak Dam demonstrates the validity and applicability of the laboratory measured K_d's for ^{90}Sr to the natural environment.

Similar calculations for ^{60}Co and ^{137}Cs would indicate that discharges at White Oak Dam of 0.5 and 1.2% of MPC, respectively. Since our previous work (Spalding and Cerling 1979) indicated that substantial proportions of the ^{60}Co and ^{137}Cs of streambed gravel were contained in tightly-bound and, presumedly, slowly equilibrating phases, the laboratory K_d's for these radionuclides were likely conservative, i.e., low. The predicted concentrations in the water would, therefore, be overestimated; but, as just shown, these calculated high concentrations were still far below the MPC's. This comparison emphasizes the relative importance of ^{90}Sr contamination discharging from White Oak Creek watershed even though the activity of ^{60}Co and ^{137}Cs in the sediments is much higher.

The influence of mineralogical composition on the K_d for ^{85}Sr can also be deduced from Table 1. The gravels in upper White Oak Creek in Bethel Valley were dominated by limestone, chert, and sandstone; these arise from the Chickamauga limestone and Knox dolomite and, to a lesser extent, the Rome formation, a mixture of shale, siltstone, and sandstone. In contrast, Melton Valley, which includes lower White Oak Creek and Melton Branch, is underlain by Conasauga shale and, hence, these streambed gravels tend to be dominantly shale fragments. The first five samples in Table 1 (from upper White Oak Creek) had K_d's for ^{85}Sr of 20 ml/g or less. The five samples from lower White Oak Creek had an average Kd of 47.5 ml/g which likely reflects the increasing proportion of shale in these gravels. The average K_d for gravels from streambeds, originating wholly in Melton Valley and consisting almost exclusively of shale fragments, was 89 ml/g. Gravels

which contained only limestone and chert were previously observed to have K_d's of 6 and 20 ml/g, respectively (Spalding and Cerling 1979). A mixture of low K_d limestone and chert with higher K_d shale gravel in White Oak Creek as it flows through Melton Valley explains the generally increasing K_d of the gravels downstream (Table 1, first ten samples). The four gravels from the Northwest tributary, composed mainly of chert, exhibited an average K_d of 30 ml/g. Such differences in the K_d of different sediments must be used to attenuate the interpretation of ^{90}Sr concentrations of gravels when comparing different drainage basins within White Oak Creek watershed.

Areal Distribution of Radionuclides in White Oak Creek Basin

There was a continuum of radioactive contamination present in White Oak Creek watershed gravels from background levels to over 10,000 dpm/g for ^{60}Co and ^{137}Cs and 1000 dpm/g for ^{90}Sr. Background levels, which, for ^{90}Sr and ^{137}Cs, were due primarily to atmospheric fallout but include the detection limits quoted below, were estimated to be 1.0, 2.0, and 2.0 dpm/g for ^{60}Co, ^{90}Sr, and ^{137}Cs, respectively. The frequency distributions of each radionuclide concentration in the lower activity ranges are depicted in Fig. 4 which provided the criteria for the selection of the above background levels. These background levels included counting uncertainties, which were functions of counting times, background counting rates, and efficiencies; detection limits were estimated to be 1.0, 0.7, and 1.0 dpm/g for ^{60}Co, ^{90}Sr, and ^{137}Cs, respectively.

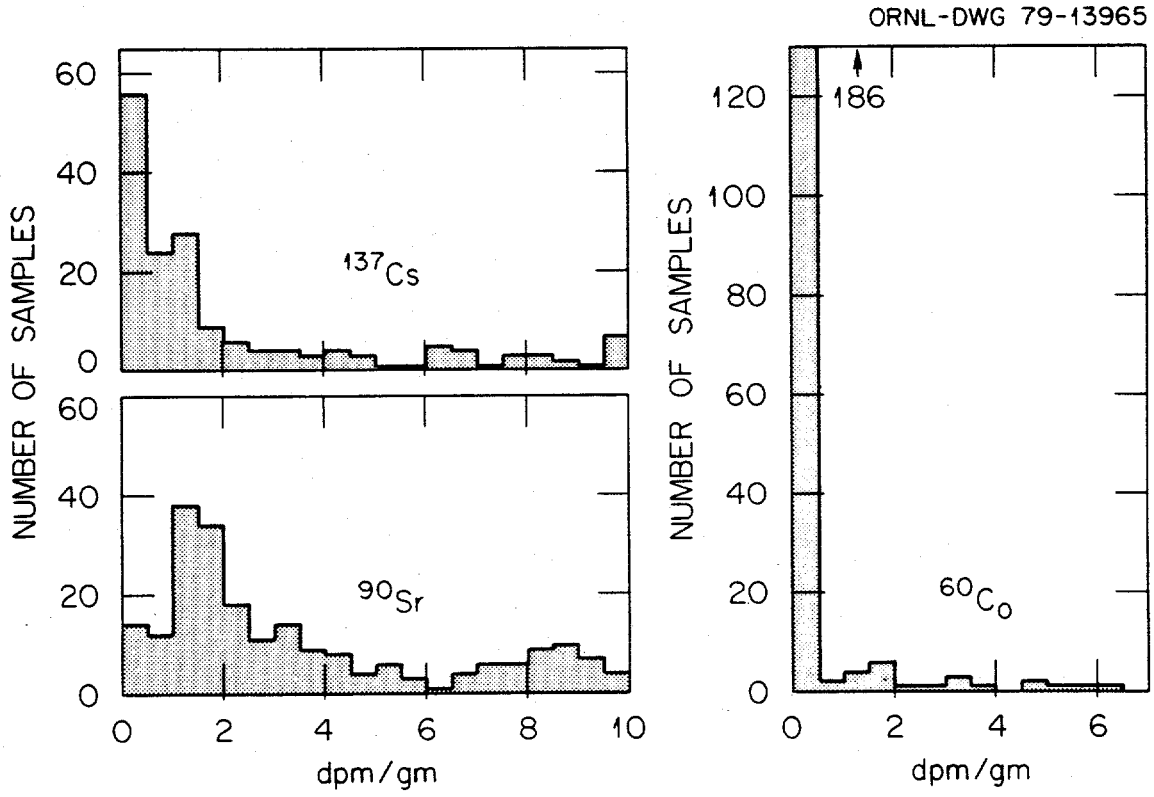

Fig. 4. Frequency distributions of ^{137}Cs, ^{90}Sr, and ^{60}Co in streambed gravels of White Oak Creek watershed at the lower activity ranges.

The gravels from each stream contained coatings of hydrous oxides of Fe and Mn which have been shown to be quite important in the adsorption and immobilization of ^{60}Co (Means et al. 1978, Spalding and Cerling 1979). Since they also play a role in the adsorption of ^{90}Sr and ^{137}Cs, the amounts of Fe and Mn in the gravel extracts, prepared for ^{90}Sr determination, were also measured. The frequency distribution for the concentration of each in the 412 samples collected are presented in Fig. 5. Although we will but briefly discuss these Fe and Mn concentrations of these gravels in this report, we present these concentrations in Fig. 5 and in the subsequent streambed concentration profiles to enable the reader to compare White Oak Creek sediments with those from other watersheds.

Obviously, the distribution of radionuclide concentrations can be used directly to locate sources of contamination entering White Oak Creek watershed; this will be discussed subsequently. However, to interpret these concentrations in terms of their relative contributions to the total radionuclide discharge from the watershed, necessitates some further considerations. Firstly, a mean concentration of the radionuclide in the streamwater in contact with the gravel must be calculated; the K_d values, discussed previously, serve quite well for these calculations. Although the absolute values of these K_d's may be, at worst, unrealistically low, their relative values for comparing different gravels should be quite useful. As discussed previously and subsequently, the values of the ^{90}Sr K_d's appeared to be quite realistic since the calculated water concentrations agree closely with the measured concentrations at various monitoring stations in the

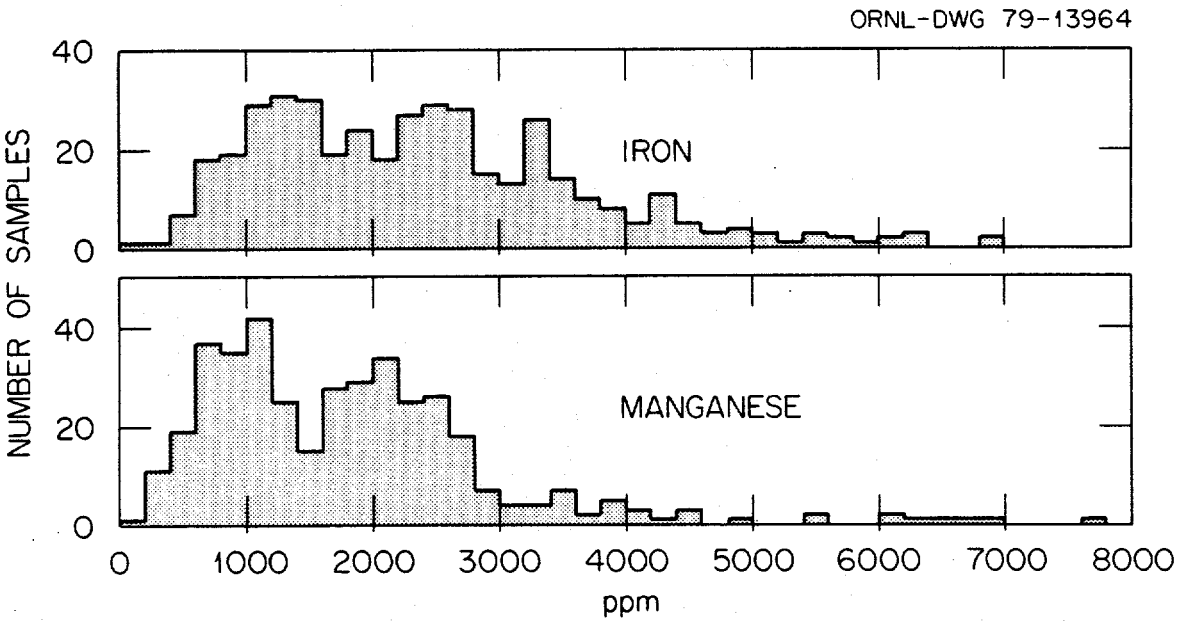

Fig. 5. Frequency distribution of hydroxylamine-extractable Fe and Mn of streambed gravels in White Oak Creek watershed.

watershed. The K_d values for ^{60}Co and ^{137}Cs may be low and, hence, lead to overestimates of their water concentrations.

Secondly, the water discharges at the locations where the gravels were sampled must be estimated. The total annual volume of water flowing past this location multiplied by the average concentration would yield the total discharge at that point. If it is assumed that the annual water discharge at a particular location is proportional to the area of land drained above that location, then the relative contribution of a particular stream or location can be estimated by multiplying the mean water concentration by the area drained. Stream discharges can vary substantially in their contributions from surface runoff and groundwater. Nevertheless, total discharge, whether it originates from surface runoff or groundwater, would still be proportional to drainage area given the similarity of topography, vegetation, and soils within the relatively small area of White Oak Creek watershed. However, substantial amounts of water are imported into the watershed from the Clinch River for the cooling of several reactors at the laboratory; such a condition would lead to an underestimation of stream discharge via drainage area for those drainages receiving substantial amounts of imported water. Nevertheless, we could have estimated radionuclide discharge in becquerels using an empirically determined water discharge to drainage area ratio multiplied by the calculated radionuclide concentrations in the water. We chose not to calculate such radionuclide discharges but rather to use radionuclide concentrations and drainage areas to construct a ranking system of the problem areas within the watershed.

To perform this analysis, White Oak Creek drainage basin was divided into several subbasins (Fig. 2); these smaller component drainages were chosen based on the areal distributions of the radionuclides; uncontaminated sections were grouped together unless they contributed to the discharge of a contaminated reach. The areas of the drainages in Fig. 2 were measured by planimetry of the topographic map of the watershed and are presented in Table 2.

For each drainage, a concern factor (CF) was defined:

$$CF = \frac{(\text{mean gravel radionuclide concentration}) \times (\text{drainage area})}{K_d}$$

The mean radionuclide concentration of the gravel was computed for each drainage using, where possible, ten samples immediately upstream from the point of interest (usually the mouth). Several streams contained fewer than ten samples below sources of contamination; these upstream uncontaminated samples were not included in these means since the purpose was to produce the best estimate of the water concentration at the mouth of each stream or drainage. The mean concentrations of ^{90}Sr, ^{60}Co, and ^{137}Cs in gravels from the mouths of the various drainages are presented in Table 3.

Using these mean concentrations and the empirical K_d values from Table 1, concern factors for the relative importance of each drainage to the total discharge from the watershed can be calculated. Table 4 lists the concern factors for ^{90}Sr from the various drainages in decreasing order of estimated discharge. The validity of interpreting these concern factors as relative estimates of the ^{90}Sr discharge

Table 2. Areas of subdrainages of White Oak Creek watershed important in radionuclide discharge

Subdrainage[a]	Area (hectares)
T7	2.5
OLD WOC	5.3
BG6	8.2
P234	9.6
T567	14.4
BG4	26.8
BG5E	35.6
BG6E	65.1
HFIR	71.9
NWT	175.9
MB[b]	286.0
Upper WOC[c]	667.3
Lower WOC[d]	202.0
Total	1570.6

[a]See Fig. 2 for description of subdrainages.

[b]Above monitoring station 4 but excluding BG5E and HFIR.

[c]Above monitoring station 2A but excluding NWT.

[d]Below monitoring station 2A excluding all subdrainages listed.

Table 3. Mean streambed gravel concentration of ^{90}Sr, ^{60}Co, and ^{137}Cs in the mouths of various drainages within White Oak Creek watershed

Subdrainage[a]	Samples used for calculation[b]	Mean activity-dpm/g		
		^{90}Sr	^{60}Co	^{137}Cs
P234	295 to 306	3.2	963	25
BG6	344 to 350	172.8	1	1
T7	243 to 245	4.3	22,000	87
T567	321, 322, 323, 325 330 to 331	6.9	206	33
OLD WOC	246 to 256 109, 110	72.6	158	4,380
BG5E	197 to 206	107.2	1	2,110
HFIR	174 to 177	2.4	1,533	11
BG4	94 to 108	1,077.0	6	146
BG6E	231 to 242	38.4	343	8
NWT	282 to 290	5.8	1	1
MB[c]	147 to 156	37.4	1,180	217
Upper WOC[d]	27, 28, 29, 31, 32, 33, 35, 36, 38, 39	10.0	130	3,460[e]
Lower WOC[f]	84 to 93	32.5	389	5,745
Upper WOC[g]	59 to 67, 373	15.4	150	1,970
Below wod	335 to 340	31.1	420	8,690

[a]See Fig. 2 for descriptions of subdrainages.

[b]See Appendix for sample location and description.

[c]Above monitoring station 4.

[d]Above monitoring station 2A.

[e]Without sample 32 which was anomalously high in ^{137}Cs (23,200 dpm/g).

[f]Immediately above White Oak Lake.

[g]Above monitoring station 3.

Table 4. Contributions of subdrainages to the total ^{90}Sr discharge discharge from White Oak Creek watershed

Subdrainage[a]	Mean ^{90}Sr[b] (dpm/g)	Estimated K_d (ml/g)	Drainage area (ha)	^{90}Sr Concern[c] factor
WOC (above MS 3)	15.4	17[d]	875.3	793
WOC (above MS2A)	10.0	17[d]	843.2	496
BG4	1077.0	89[e]	26.8	324
MB (above MS4)	37.4	89[e]	393.5	165
BG5E	107.2	89[e]	35.6	43
NWT	5.8	30[f]	175.9	34
BG6E	38.4	89[e]	65.1	28
BG6	172.8	89[e]	8.2	16
OLD WOC	72.6	25[g]	5.3	15
HFIR	2.4	89[e]	71.9	2
T567	6.9	89[e]	14.4	1
P234	3.2	89[e]	9.6	0.3
T7	4.3	89[e]	2.5	0.01
Above wol[h]	32.5	47[i]	1402.9	970

[a]See Fig. 2 for description of subdrainages.

[b]Values for the subdrainages were taken from Table 3.

[c]Concern Factor = (Mean ^{90}Sr / K_d) x Drainage Area.

[d]Mean of samples 1, 11, 21, 31, and 41 (Table 1).

[e]Mean of samples 100, 150, 175, 205, 240, 305, 330, and 350 (Table 1).

[f]Mean of samples 260, 270, 280, and 290 (Table 1).

[g]Sample 250 (Table 1).

[h]All drainage area above the mouth of White Oak Creek at its discharge into White Oak Lake.

[i]Mean of samples 51, 61, 71, 81, and 91 (Table 1).

from these drainages can be checked in several ways. For instance, the ratio of concern factors for White Oak Creek above monitoring station 3 and Melton branch above monitoring station 4 computed to 4.8. The average ratio of the monitored ^{90}Sr discharge at these two stations for the twelve months prior to our sampling (October, 1978) was 3.7; the monthly ratio varied from 9.7 to 0.8 (Lasher 1977, 1978). This would indicate that the concern factors ratio yielded a realistic description of the relative contributions of these two major drainages. As a further check, these two concern factors appeared to add up to the concern factor for White Oak Creek as it entered White Oak Lake, i.e., 793 + 165 = 958 versus the calculated 970 (Table 4). Notably, the difference between concern factors above monitoring station 3 (793) and that above monitoring station 2A(496) was 297; the calculated concern factor for the two drainages (BG4 and old WOC, Fig. 2) discharging between these points was 324 + 15 = 339. This agreement could actually be closer since part of the drainage in BG4 was diverted into the old channel of White Oak Creek at the time of our sampling. This could easily lower the BG4 concern factor by 10%. The correlative and additive attributes of these concern factors for the drainage areas whose ^{90}Sr discharges were known lend some confidence in their application to smaller drainages or subsections of larger drainages which are not routinely monitored. These same correlative and additive attributes of the concern factors also make the assumptions concerning the drainage area-discharge proportionality appear valid.

As judged by the concern factors in Table 4, the major areas contributing to the total ^{90}Sr discharge were located somewhere above monitoring station 3(793 out of 970 CF or 82%). This contribution can be subdivided further based on the CF's in Table 4. The old channel of White Oak Creek with its contaminated floodplain and the drainage from SWDA4 contributed 324/970, or 33%. The Northwest Tributary contributed 34/970 or 3.5%; and 496-34/970 or 48% arose from points above monitoring station 2A, but excluding the Northwest Tributary. This 48% originated from ORNL plant effluents; this conclusion will be substantiated subsequently when the areal distribution of the ^{90}Sr concentrations are presented. An additional fraction (165/970 or 17%) was contributed by Melton branch and, presumedly, due mainly to SWDA 5 leaching directly into this stream; the only other contaminated sources contributing to the Melton branch discharge were the drainage east of SWDA 5 (BG5E, Fig. 2) and the drainage through the High-Flux Isotope Reactor complex (HFIR) which, when combined, contributed only 4.6% (i.e., 43 + 2/970) to the discharge of the watershed. Notably, two other areas, the drainages of SWDA 6 (BG6) and that east of SWDA 6 (BG6E), contributed small increments to the total discharge: 1.6%(16/970) and 2.9%(28/970), respectively. These areas are not routinely monitored and these calculated contributions to the total discharge show that they need not be at the present time; annual surveys should suffice for these and other minor sources within the watershed as listed in Table 4.

A similar calculation of the concern factors for ^{60}Co and ^{137}Cs in these same subdrainages is presented in Tables 5 and 6,

Table 5. Contributions of subdrainages to the total ^{60}Co discharge from White Oak Creek watershed

Subdrainage[a]	Mean ^{60}Co[b] (dpm/g)	Estimated K_d (ml/g)	Drainage area (ha)	^{60}Co concern[c] factor
MB (above MS4)	1,180	728[d]	393.5	638
WOC (above MS3)	150	448[e]	875.3	293
WOC (above MS2A)	130	448[e]	843.2	245
HFIR	1,533	728[d]	71.9	151
T7	22,000	728[d]	2.5	76
BG6E	343	728[d]	65.1	31
P234	963	728[d]	9.6	13
T567	206	728[d]	14.4	4
OLD WCC	158	448[e]	5.3	2
NWT	1	387[f]	175.9	0.5
BG4	6	728[d]	26.8	0.2
BG5E	1	728[d]	35.6	0.05
BG6	1	728[d]	8.2	0.01
Above wol	389	642[g]	1402.9	850

[a]See Fig. 2 for description of subdrainages.

[b]Values for the subdrainages were taken from Table 3.

[c]Concern Factor = (Mean ^{60}Co / K_d) x Drainage Area.

[d]Mean of samples 100, 150, 175, 205, 240, 305, 330, and 350 (Table 1).

[e]Mean of samples 1, 11, 21, 31, 41 (Table 1).

[f]Mean of samples 260, 270, 280, and 290 (Table 1).

[g]Mean of samples 51, 61, 71, 81, and 91 (Table 1).

Table 6. Contributions of subdrainages to the total ^{137}Cs discharge from White Oak Creek watershed

Subdrainage[a]	Mean ^{137}Cs[b] (dpm/g)	Estimated K_d (ml/g)	Drainage area (ha)	^{137}Cs concern[c] factor
WOC (above MS2A)	3,460	4,110[d]	843.2	710
WOC (above MS3)	1,970	4,110[d]	875.3	420
MB (above MS4)	217	12,100[e]	393.5	7.1
BG5E	2,110	12,100[e]	35.6	6.2
OLD WOC	4,380	4,110[d]	5.3	5.6
BG4	146	12,100[e]	26.8	.32
HFIR	11	12,100[e]	71.9	.07
BG6E	8	12,100[e]	65.1	.04
T567	33	12,100[e]	14.4	.04
NWT	1	5,510[f]	175.9	.03
T7	87	12,100[e]	2.5	.02
P234	25	12,100[e]	9.6	.02
BG6	1	12,100[e]	8.2	.01
Above wo1	5,745	10,700[g]	1,402.9	753

[a]See Fig. 2 for description of subdrainages.

[b]Values for the subdrainages were taken from Table 3.

[c]Concern Factor = (Mean ^{137}Cs / Kd) x Drainage Area.

[d]Mean of samples 1, 11, 21, 31, and 41 (Table 1).

[e]Mean of samples 100, 150, 175, 205, 240, 305, 330, and 350 (Table 1).

[f]Mean of samples 260, 270, 280, and 290 (Table 1).

[g]Mean of samples 51, 61, 71, 81, and 91 (Table 1).

respectively. Although the discharges at White Oak Dam are far below MPC's for both these radionuclides, the concern factors do allow the estimation of the relative contributions of different areas to their total discharge. In addition, the relative concern factors for all three radionuclides could be compared by dividing them by the MPC's for each radionuclide (i.e., 0.67, 111, and 44 dpm/ml for ^{90}Sr, ^{60}Co, and ^{137}Cs, respectively). Such a calculation for all sources above White Oak Lake yielded comparative concern factors of 1450, 7.7, and 17 for ^{90}Sr, ^{60}Co, and ^{137}Cs, respectively. This calculation further illustrates why our attention continues to be focused on ^{90}Sr.

The major burden of the ^{60}Co discharge arose in Melton Branch, i.e., 638/850 or 75%. The major source of this ^{60}Co was the cooling water drainage from the High-Flux Isotope Reactor (HFIR). Although the CF for HFIR calculated to only 151, this number was based on only four samples between the outlet of the cooling water and the confluence of the drainage with the main channel of Melton Branch. It appeared likely that the ^{60}Co had not reached equilibrium with the gravels in this short reach of stream and, hence, fell below a true equilibrium with the gravels in this short reach of stream; a plausible reason for this lack of equilibrium was the rather elevated temperature of the water in this short reach of creek (usually greater than 40°C), until it was cooled by mixing with the water in the main channel. The more conclusive evidence that this HFIR drainage was the major source of ^{60}Co can be deduced from the concentration distribution profile in this stream (to be discussed subsequently). There also exists a well-studied seep of ^{60}Co east of intermediate-level liquid waste

trench 7 (T7, Fig. 2) (Means et al. 1978). Our calculated concern factor for T7 was 76 which would be only 8.9% of the total basin concern factor of 850 (Table 5). Although the ^{60}Co concentrations in the gravels of T7 were high (22,000 dpm/g), the drainage was quite small (2.5 ha); this concern factor serves to put this well-known seep into perspective. A third drainage of concern with respect to ^{60}Co is the area above monitoring station 2A; as was observed with ^{90}Sr, most of this discharge arose from ORNL plant effluents. Other areas listed in Table 5 contributed only small increments to the total drainage discharge. Although the ^{60}Co concentrations in the gravels were sometimes high, the high K_d of these gravels for ^{60}Co, coupled with the comparatively small areas drained by these streams, led to these low concern factors.

The concern factors for ^{137}Cs (Table 6) showed that practically all of the basin's discharge originated above monitoring station 2A and, hence, from ORNL plant effluents. It should be noted that the concern factors for ^{137}Cs did not have the additive attributes as those for ^{90}Sr and ^{60}Co; the CF for ^{137}Cs actually dropped from 710 to 420 in White Oak Creek between monitoring stations 2A and 3, a region where this difference should have increased. A major source of this discrepancy might be the very high variability in the ^{137}Cs K_d values (Table 1) and, hence, the estimated K_d's (Table 6) employed in the CF calculation. Nevertheless, the high K_d's of all basin gravels for ^{137}Cs were obtained throughout the main channel of White Oak Creek. This high affinity for ^{137}Cs by the gravels and, presumedly, the soils from which they originated, is the major factor limiting the

discharge of ^{137}Cs from all waste disposal sites in the watershed. It seems pertinent to note that, due to the high K_d of the gravels for ^{137}Cs and the great difficulty in extracting ^{137}Cs from streambed gravels (Spalding and Cerling 1979), the ^{137}Cs in these gravels would have to arise from either direct discharge into the creek (such as with ORNL plant effluents) or erosion of contaminated surface soil into the creek. In addition, ^{137}Cs would likely move in the creek in a suspended particulate phase as bed sediments are weathered into smaller particles which can be more easily suspended by the creek water.

The areal distribution of ^{90}Sr concentrations in the gravels within the basin are depicted in Fig. 6. With the aid of such a map, the precise points of ^{90}Sr contamination, where they enter the various subdrainages, can be located. Identical maps showing the areal distribution of ^{60}Co and ^{137}Cs concentrations are presented in Fig. 7 and Fig. 8, respectively. It should be reiterated that high concentrations are not necessarily indicative of high discharges at these locations. What the concentrations do show is the location where contamination begins in a given reach of creek. For instance, ^{90}Sr, ^{60}Co, and ^{173}Cs concentrations in the main channel of White Oak Creek from points above ORNL plant effluents to below White Oak Dam are presented in Fig. 9. Above the ORNL plant effluents, all three radionuclides exhibited concentrations close to atmospheric fallout or background levels; in the reach of creek immediately below these effluents, the levels of ^{90}Sr, ^{60}Co, and ^{137}Cs had risen by 1, 2, and 3 orders of magnitude, respectively. These ORNL plant effluents

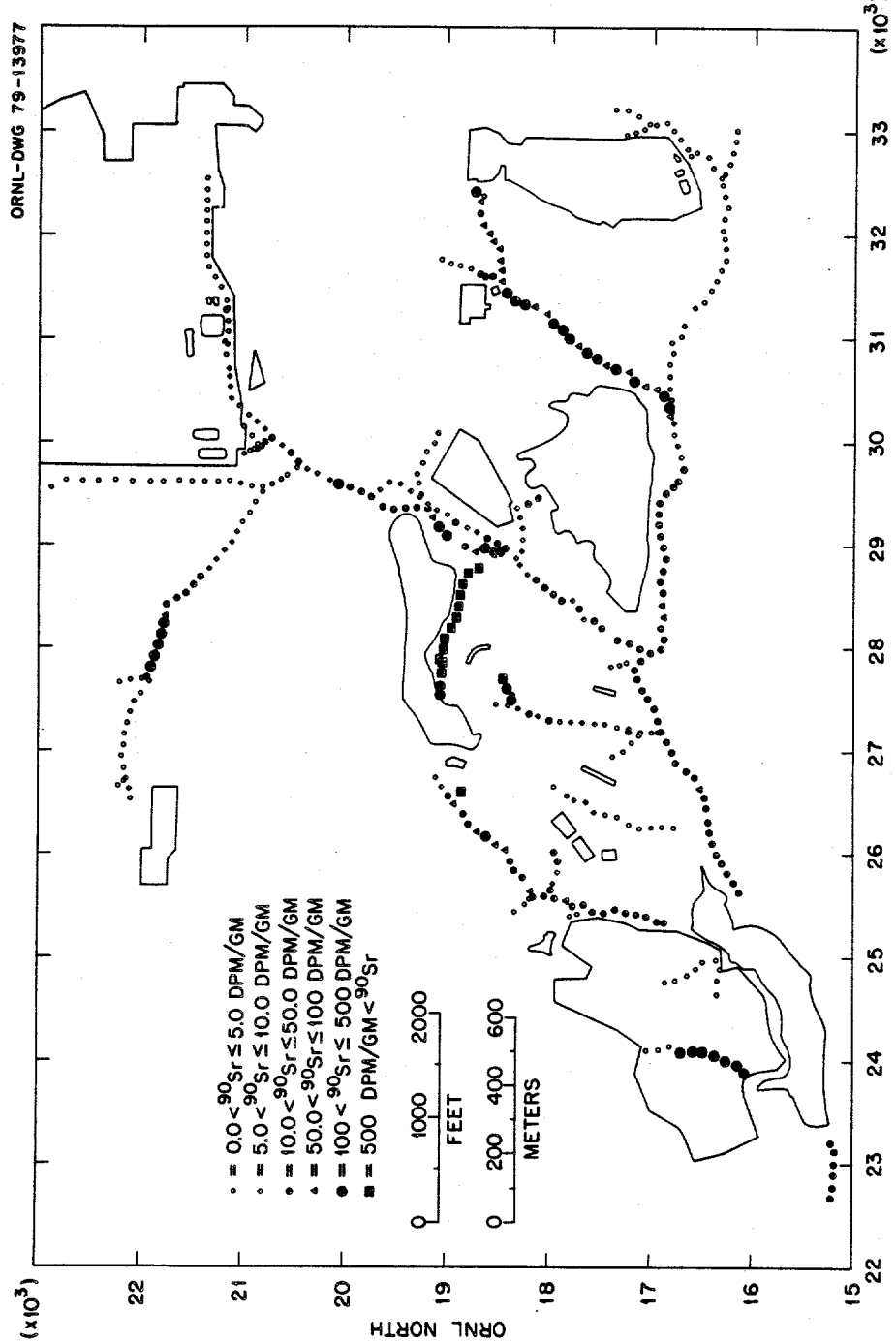

Fig. 6. Areal distribution of ^{90}Sr activity in streambed gravels of White Oak Creek watershed.

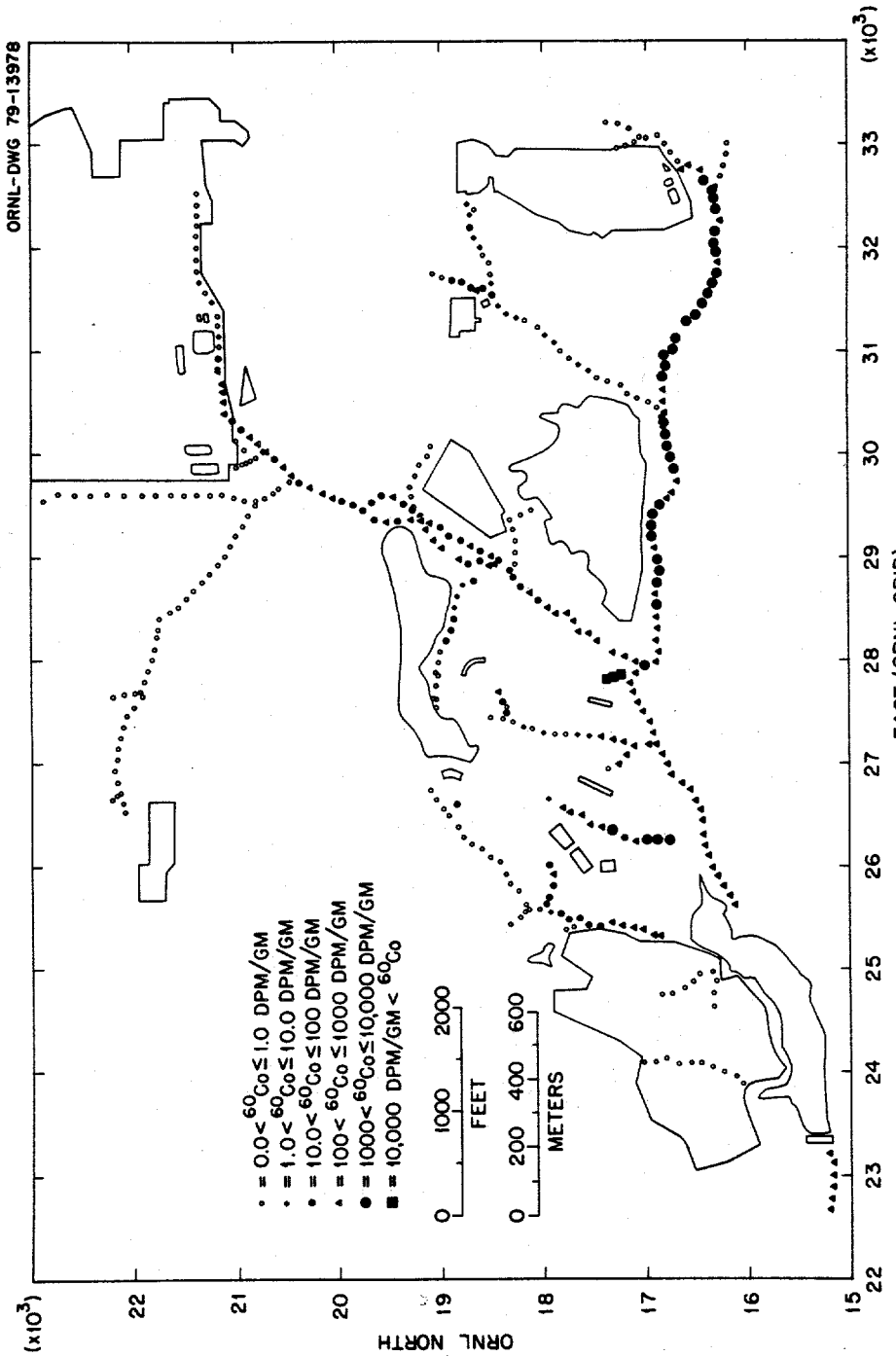

Fig. 7. Areal distribution of ^{60}Co activity in streambed gravels of White Oak Creek watershed.

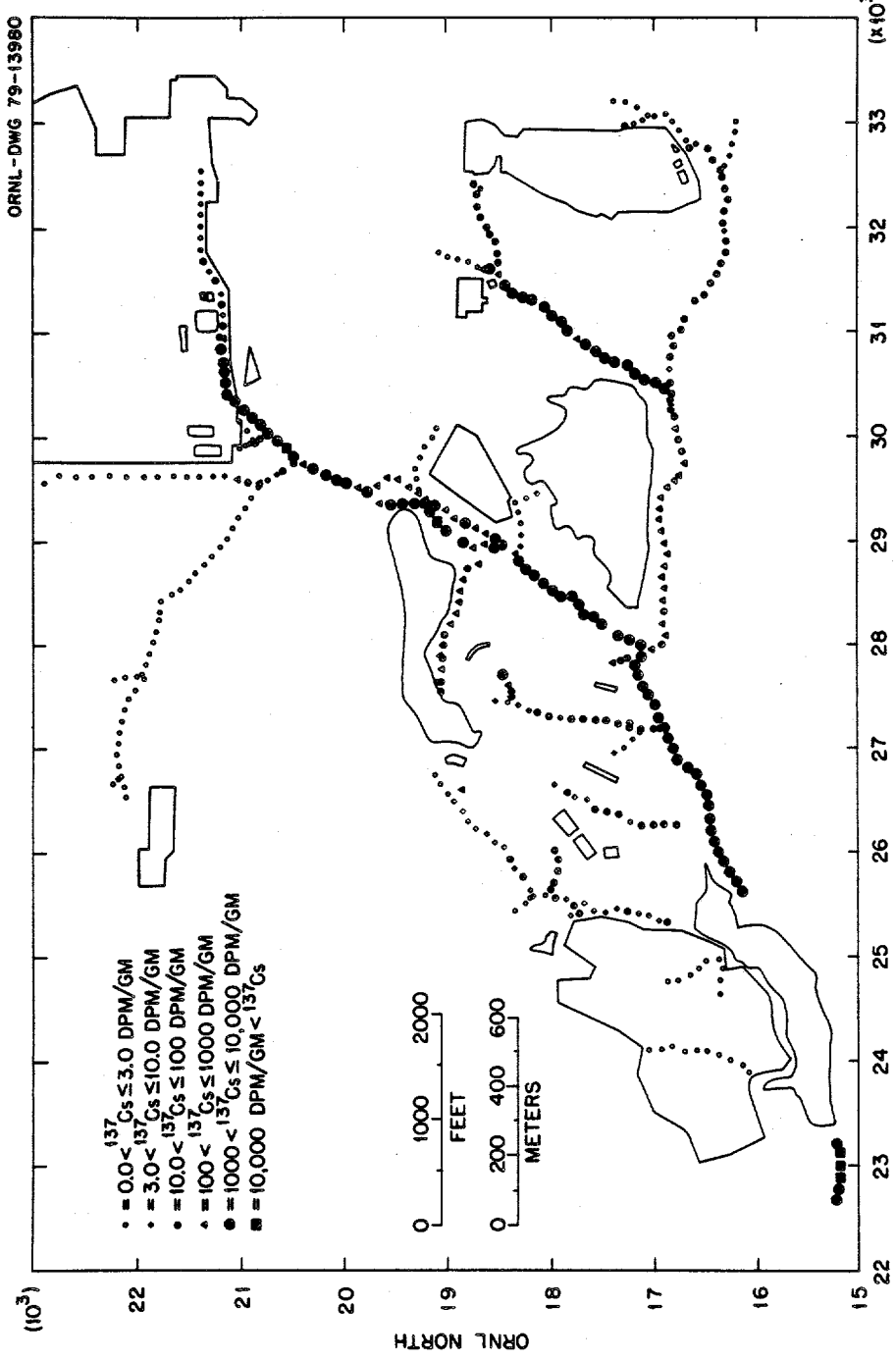

Fig. 8. Areal distribution of ^{137}Cs activity in streambed gravels of White Oak Creek watershed.

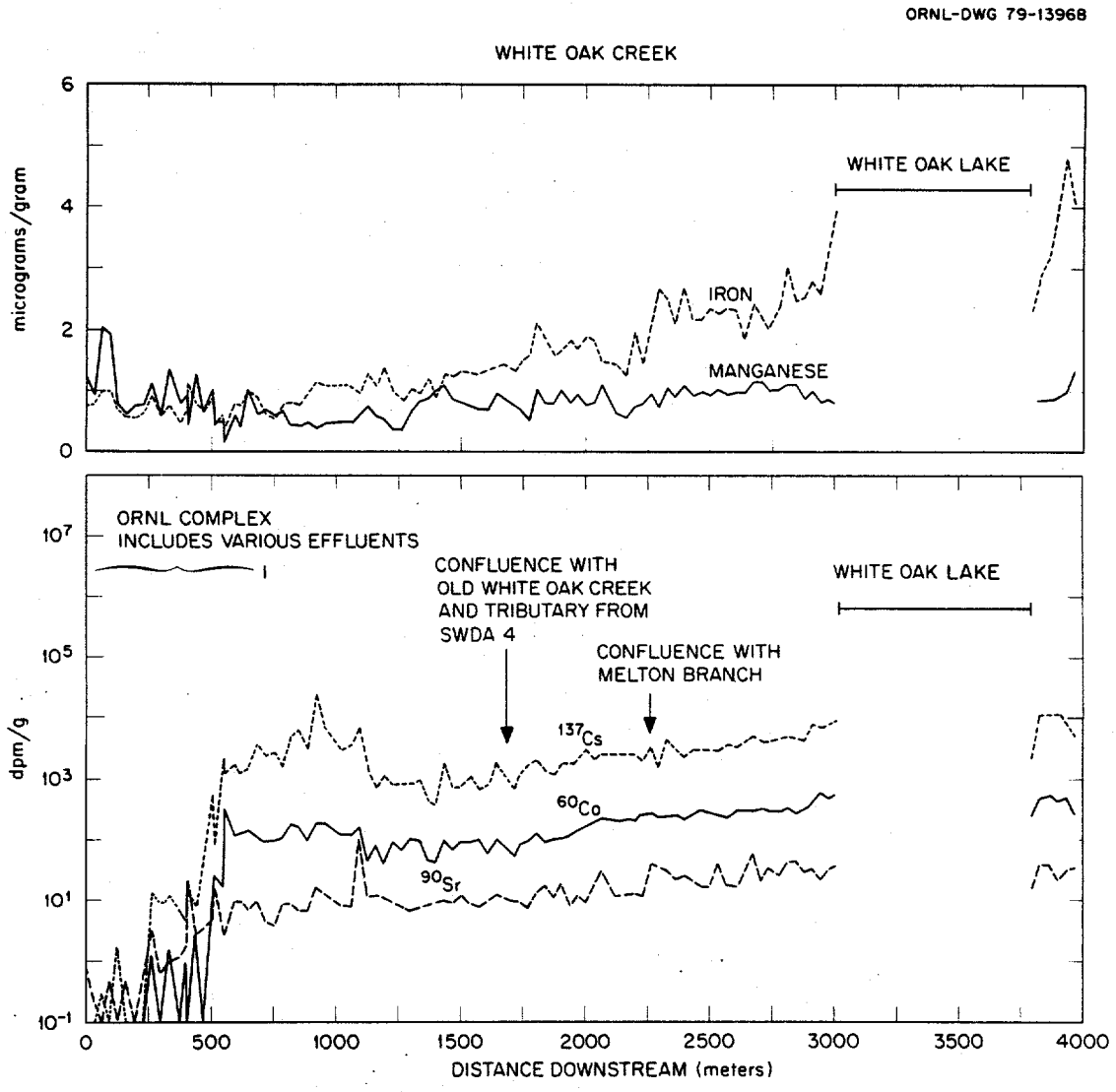

Fig. 9. Radionuclide concentration profile of White Oak Creek gravels from above ORNL plant site to below White Oak Dam.

include numerous point sources which were not differentiated here; an extended discussion of these ^{90}Sr sources can be found in the recent work of Stueber et al. (1978). The concentration of all three radionuclides in the gravels continued to rise along the entire length of White Oak Creek. The concentration of ^{90}Sr appeared to rise after the confluence of the tributary draining SWDA 4 and again after the confluence of Melton Branch. This gradual rise was most likely due to the increasing proportion of shale in the gravel fraction; as discussed previously, the K_d of White Oak Creek gravels for ^{90}Sr tended to increase downstream as the mineralogical composition changed from chert and limestone to shale. In addition, it should be noted that White Oak Lake did not appear to function as either a source or sink for any of the three radionuclides since their concentrations below the dam were equal to or slightly greater than those just above White Oak Lake.

Another area of concern for ^{90}Sr discharge was the drainage from SWDA 4 and its nearby floodplain of White Oak Creek. This area ranked second in importance to ORNL plant effluents in ^{90}Sr concern factor (Table 4). Figure 10 presents the concentration profile of the three radionuclides in these stream gravels. The ^{90}Sr concentration rose to over 1000 dpm/g near the middle of SWDA 4, i.e., about 150 m downstream from the first flowing water; this high level was maintained to its confluence with the old channel of White Oak Creek. The abrupt rise in ^{137}Cs and ^{60}Co and the fall in ^{90}Sr of this profile were due to the dilution in the last two samples which were actually in this old channel of White Oak Creek. The gradual rise in ^{90}Sr concentration within this stream would indicate the diffuse nature of the ^{90}Sr sources leaching into the creek from SWDA 4.

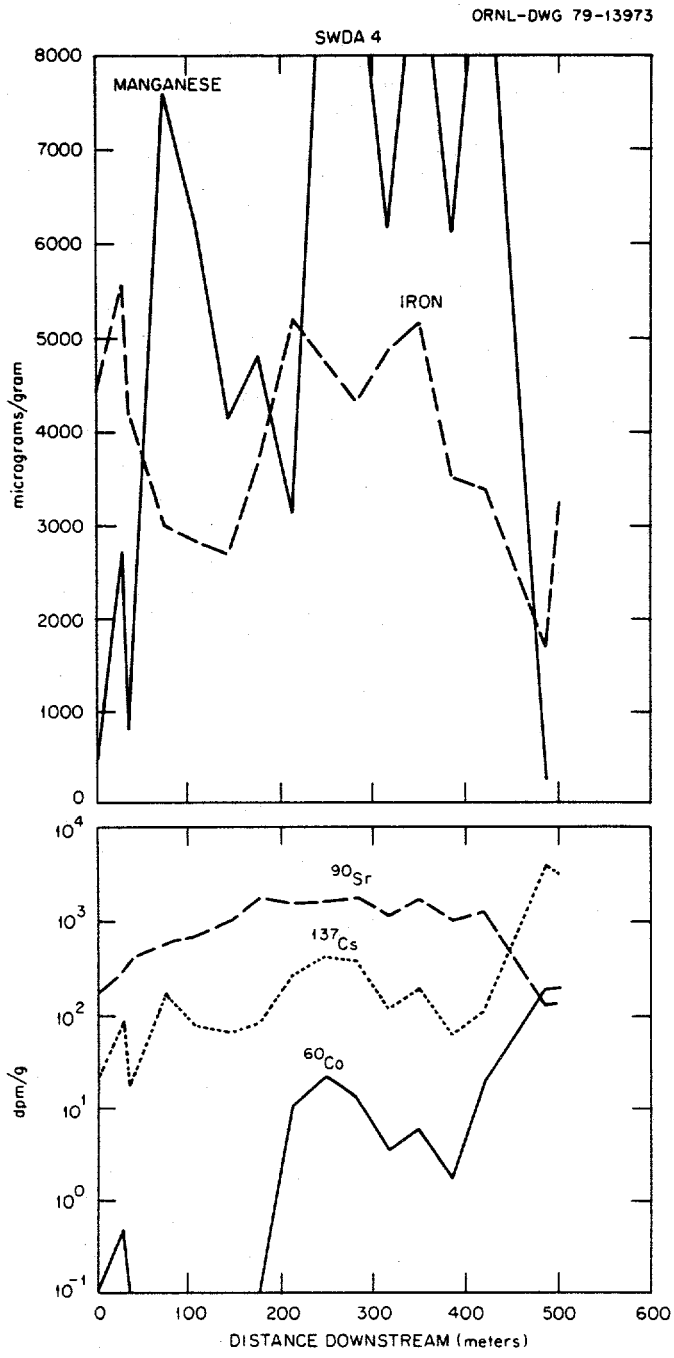

Fig. 10. Radionuclide concentration profile of gravels in stream south of SWDA4 to its confluence with the main channel of White Oak Creek.

Melton Branch, on the other hand, exhibited some marked point sources of radionuclide contamination. The concentration profile depicted in Fig. 11 started at the fartherest upstream sampling point in the stream east of HFIR (Fig. 1) and followed the creek downstream to below White Oak Dam. The most salient point source of contamination was the sharp rise in ^{60}Co level at the confluence with the HFIR cooling water effluent. This high concentration was maintained downstream until a major dilution occurred at its confluence with the main channel of White Oak Creek. This source represented the major source of ^{60}Co in the whole watershed as discussed previously. The concentration profile of ^{90}Sr exhibited two major sources entering Melton Branch. There was a noticeable rise in ^{90}Sr level in the creek at its confluence with the stream draining to the east of SWDA 5. More importantly, there was a gradual rise in ^{90}Sr concentration in the reach of Melton Branch south of SWDA 5; this high concentration was maintained until the confluence with the main channel of White Oak Creek. This diffuse source appeared to be due to groundwater seepage from the south side of SWDA 5 directly into Melton Branch. The initial pulse of ^{90}Sr, introduced into Melton Branch at the confluence with the stream east of SWDA 5, can be traced upstream to the region draining the area near the Molten Salt Reactor Experiment (MSRE) building (Fig. 1). Further downstream at the HRT settling basin (Fig. 1), ^{137}Cs contamination entered this stream and its concentration did not decrease significantly in this profile until its confluence with the main channel of Melton Branch (Fig. 12). Significant ^{60}Co contamination did not appear in this profile until

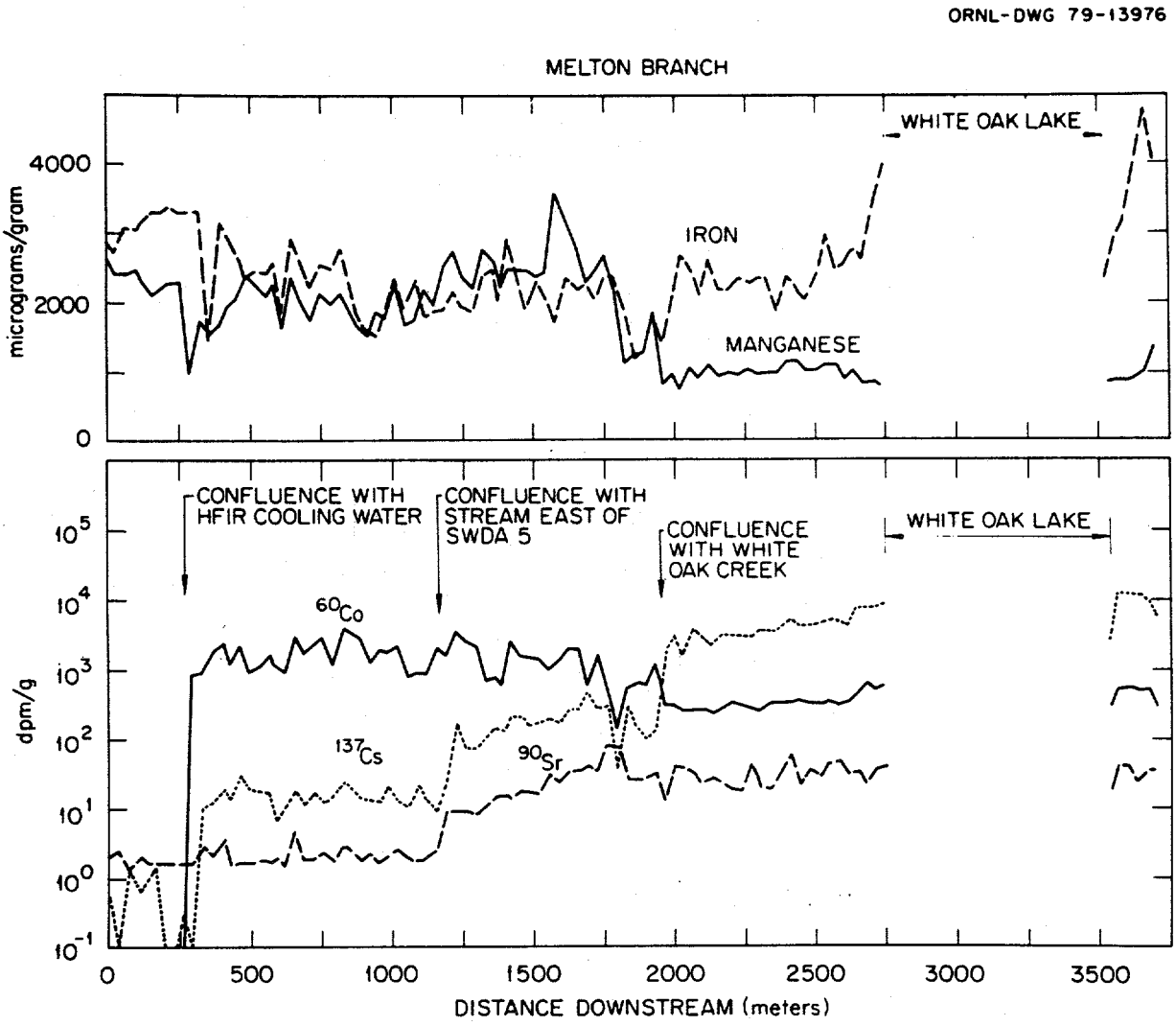

Fig. 11. Radionuclide concentration profile of gravels in Melton Branch from above HFIR cooling water effluent to below White Oak Dam.

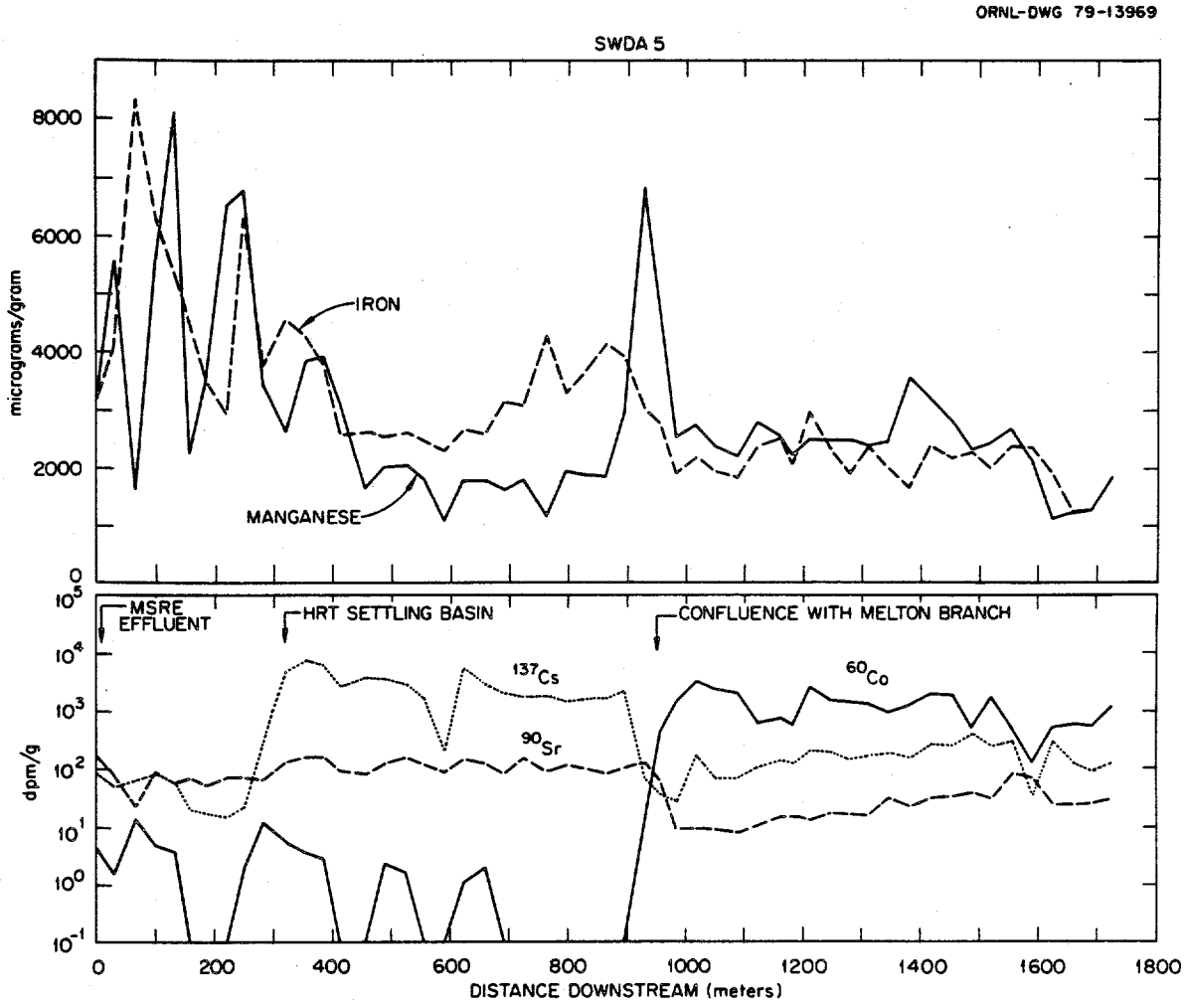

Fig. 12. Radionuclide concentration profile of gravels in the stream east of SWDA5 through Melton Branch to the main channel of White Oak Creek.

the main channel of Melton Branch; as discussed above, the HFIR cooling water was the source of this ^{60}Co. It should also be noted that the ^{90}Sr discharge from this stream east of SWDA 5 was estimated to contribute less than 5% to the total basin discharge; however, since it appeared to originate from a point source, it might be fairly easy to correct.

The Northwest Tributary (NWT) of White Oak Creek draining SWDA 3 (Fig. 1) was estimated to contribute less than 5% to the total discharge of ^{90}Sr from the whole basin (Table 4). However, most of this ^{90}Sr appeared to enter this stream at a point source (Fig. 13); this high concentration continued downstream in the gravels until the confluence with a second tributary which effected its dilution. Other work suggests that the Northwest Tributary intersects a dipping limestone which serves as a lateral aquifer carrying contaminated water from SWDA 3 (Stueber et al., in press). Other radionuclides appeared to be at or near background levels throughout the entire reach of NWT; ^{137}Cs increased just above the mouth of the NWT at its confluence with the main channel of White Oak Creek and likely represented the remnants of some floodplain or backwash from the more highly ^{137}Cs-contaminated streambed in this main channel. The drainage east of SWDA 6 was calculated to be of similar magnitude in ^{90}Sr discharge to that of the NWT. This contamination also appeared to arise in a point source from intermediate level liquid waste pit 1 (Fig. 1) and a constant level of ^{90}Sr was observed downstream (Fig. 14). In addition, about 500 m downstream two point sources of ^{60}Co were observed apparently originating from intermediate level liquid waste

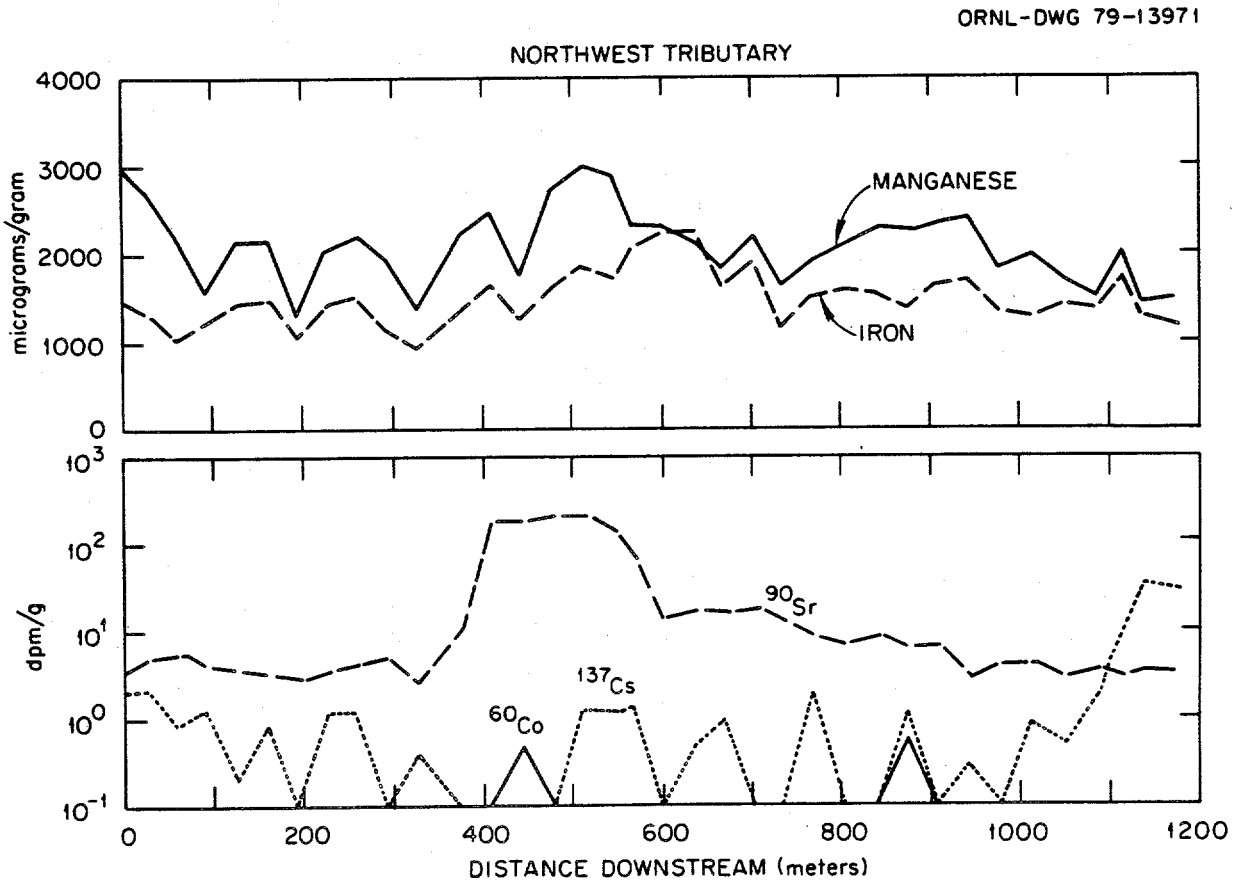

Fig. 13. Radionuclide concentration profile of gravels in the Northwest Tributary to its confluence with the main channel of White Oak Creek.

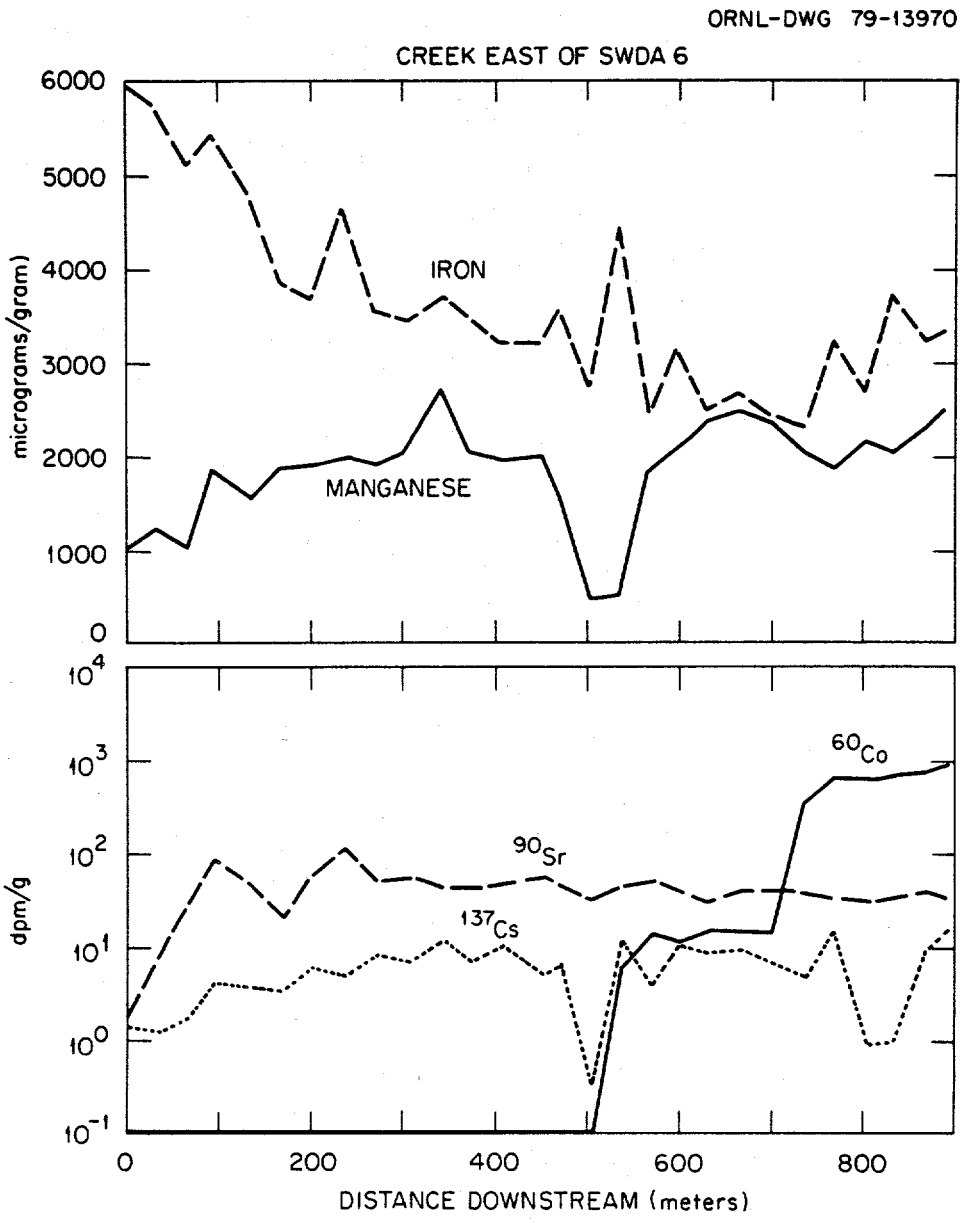

Fig. 14. Radionuclide concentration profile of gravels in the stream east of SWDA6 to its discharge into White Oak Lake.

pits 2, 3, and/or 4. It should be noted that this drainage is not presently monitored for radionuclide discharge and drains directly into White Oak Lake. However, its estimated discharge for all radionuclides would seem to preclude the necessity for routine monitoring.

A minor contribution to the total ^{90}Sr discharge also appeared to originate in the stream draining directly into White Oak Lake from the central region of SWDA 6 (Fig. 6). This contamination arose at a point source (Fig. 15); it originated as a groundwater seep emanating from a suite of recently used burial trenches filled with low-level solid waste (ca. 1973-74). This comparatively rapid migration of ^{90}Sr with groundwater illustrates the most poignant problem, the ready leachability of ^{90}Sr from the solid waste disposal areas. Remedial measures such as chemical amendment or grouting might be effective in correcting this point source. However, due to the low calculated contribution to the total discharge from this area, this ^{90}Sr contamination should probably be monitored only periodically, i.e., twice a year, to determine if its magnitude changes.

The intermediate-level liquid waste pits and trenches appeared to be minor sources of ^{60}Co contamination. Figure 16 shows the concentration profile of ^{60}Co in the stream draining pits 2, 3, 4 and trench 5. A similar profile was observed (Fig. 17) in the stream draining trenches 5, 6, and 7; here, the ^{60}Co appeared to arise from a groundwater seep on the west side of trench 7. The larger ^{60}Co seep on the east side of trench 7 (Fig. 7) appeared to be the major source of ^{60}Co from all the pits and trenches. Figure 17 also illustrates the rapid dilution of both ^{90}Sr and ^{137}Cs when the

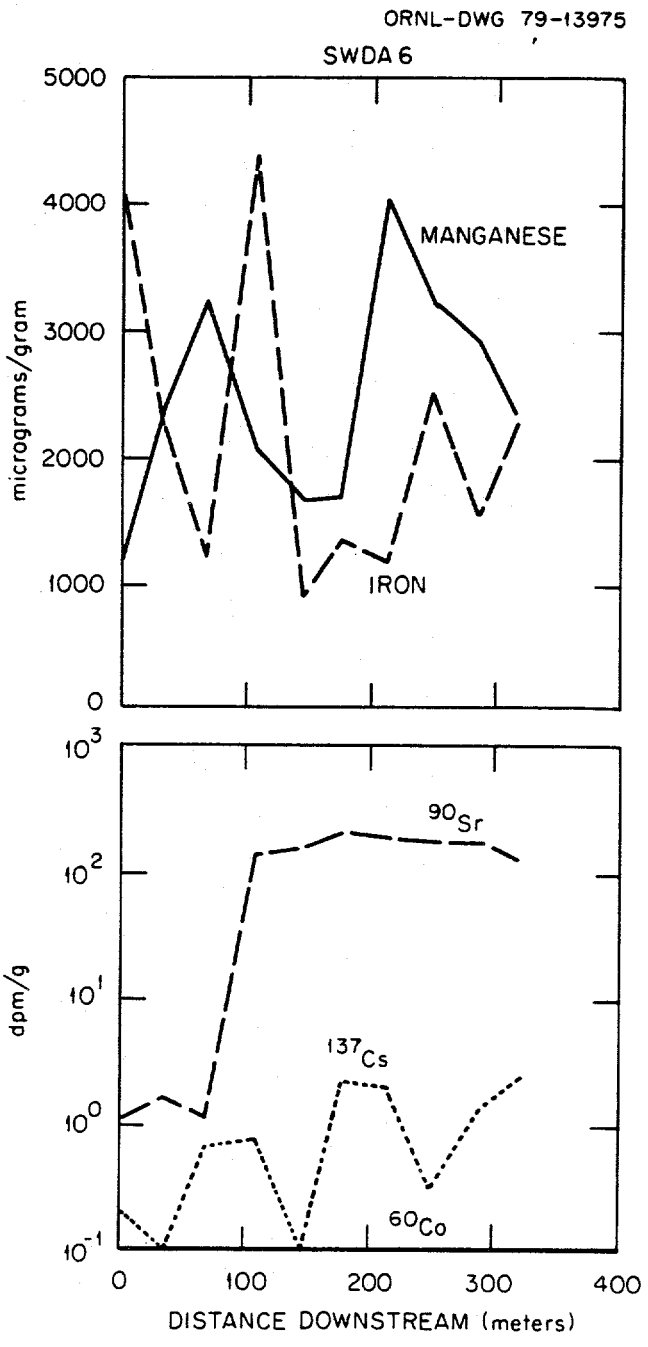

Fig. 15. Radionuclide concentration profile of gravels in the stream draining the central portion of SWDA6.

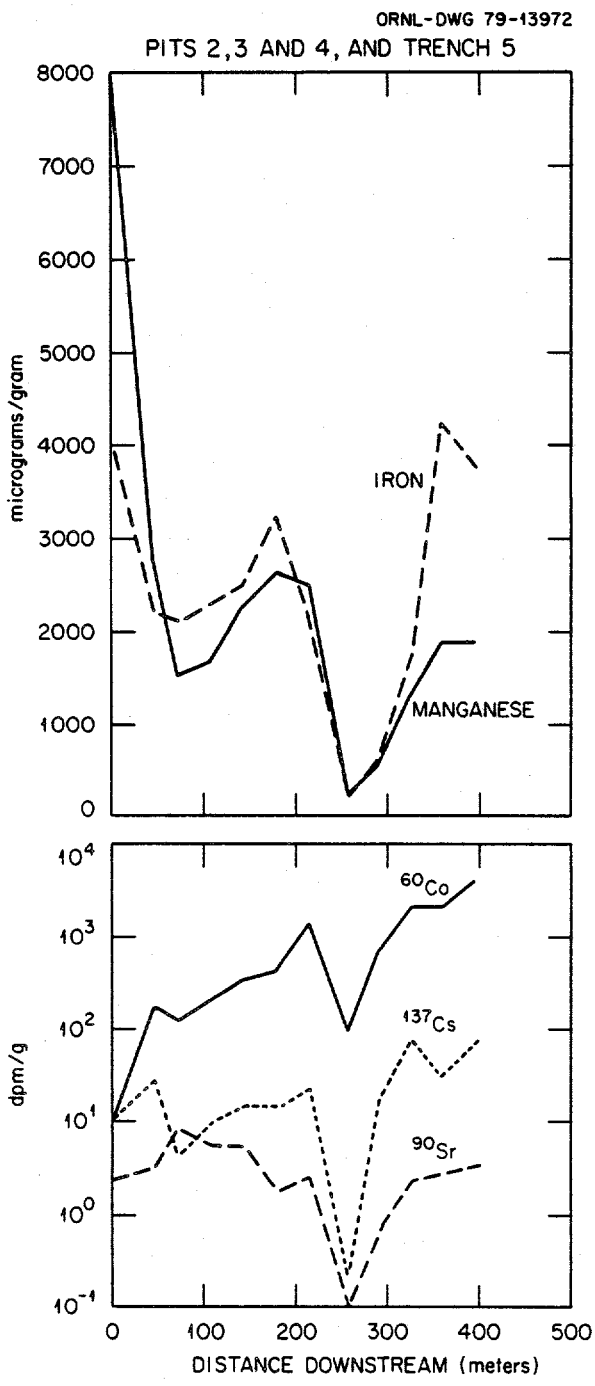

Fig. 16. Radionuclide concentration profile of gravels in the stream draining the area surrounded by liquid waste pits 2, 3, and 4 and trench 5.

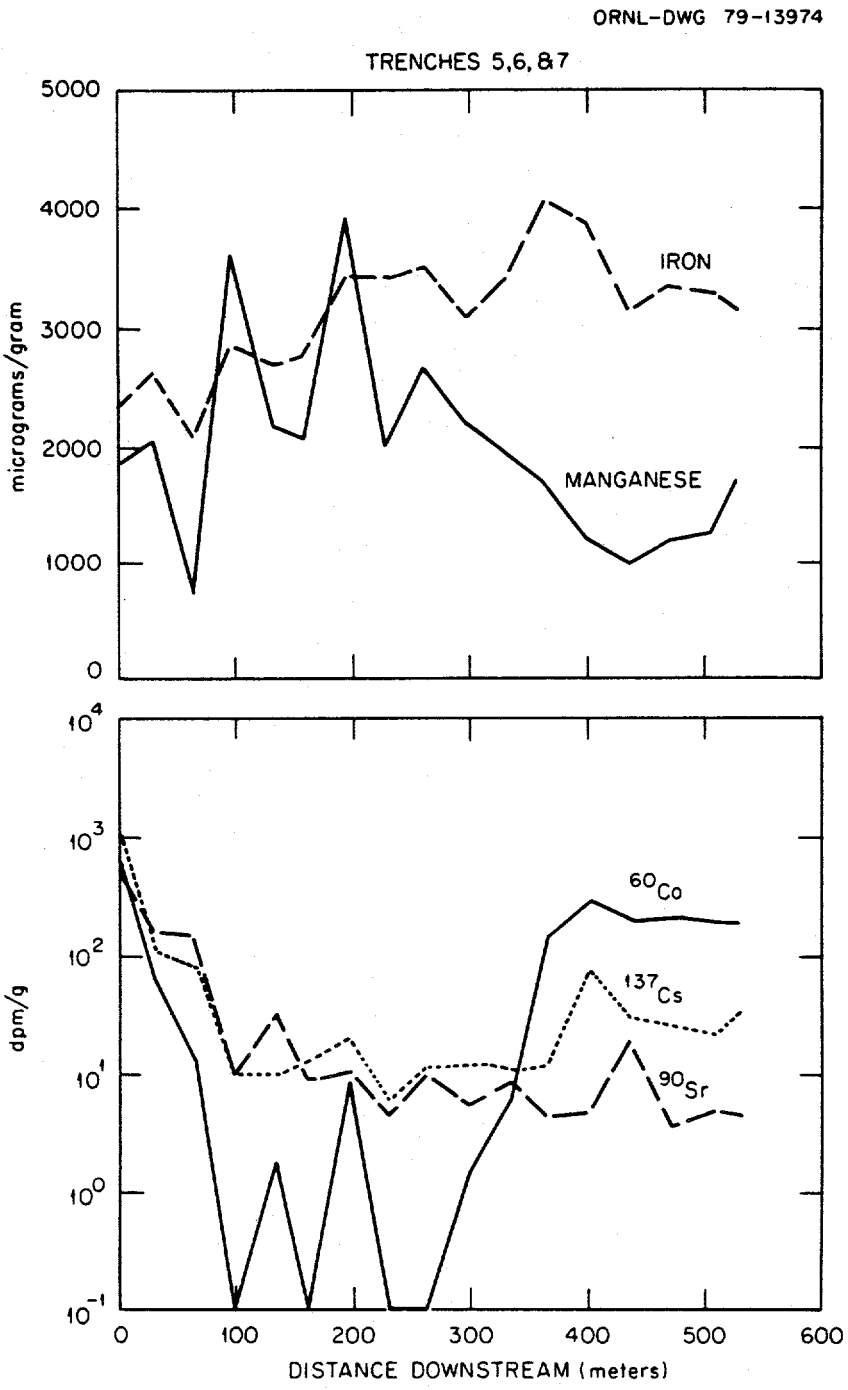

Fig. 17. Radionuclide concentration profile of gravels in the stream draining the area surrounded by liquid waste trenches 5, 6, and 7.

intermittent tributary draining from trench 6 was diluted by its confluence with the larger stream.

CONCLUSIONS

The major conclusion from this study was that radiochemical analysis of streambed gravels represents a very convenient method to precisely locate sources of radioactive contamination within a comparatively small watershed. In addition, the relative contributions from the many diverse sources in White Oak Creek watershed to the total discharge could be estimated from concentrations of radionuclides in the gravels. This comparative ranking of sources from subdivisions within the watershed was achieved with the aid of laboratory-measured K_d values for each radionuclide for each type of gravel in a particular subdrainage coupled with an assumption that the annual water flux from a given drainage was proportional to the area of that drainage. Such discharge rankings agreed with the known ^{90}Sr discharges from the monitored areas within the watershed. This agreement increased the confidence with which this ranking procedure could be applied to unmonitored drainages and subsections of the monitored drainages. This study also represents the first basin-wide survey for all sources of contamination within the White Oak Creek watershed.

The major sources of ^{90}Sr, in decreasing order on concern, were direct ORNL plant effluents, SWDA 4 with its associated contaminated floodplain of White Oak Creek, and SWDA 5 from its south side draining into Melton Branch. These three sources together were estimated to

contribute 90% of the ^{90}Sr discharge from the watershed at the time of our sampling; the individual contributions were estimated to be 50, 30, and 10%, respectively, for these three sources. Intermediate sources of ^{90}Sr included the Northwest Tributary draining SWDA 3, the tributary draining the east side of SWDA 5, and the tributary draining east of SWDA 6 with each contributing 4% or less to the total discharge. A minor source, estimated to contribute less than 2% of the ^{90}Sr discharge, was found in SWDA 6 but was of special concern since it had come about within four years of the time of waste burial.

In a more general sense, this survey showed the patterns of radionuclide contamination behavior in streambed gravels of this and similar small watersheds. There were two general types of radionuclide contamination sources; point and diffuse. Point sources yielded two types of downstream concentration profiles (Fig. 18). For instance ^{90}Sr emanating from a groundwater seep in SWDA 6 caused constant high concentrations in the gravels for 200 m downstream; this stream was not diluted by other tributaries before it discharged into White Oak Lake. A similar constant concentration profile downstream from a point source was observed from waste pit 1 (Fig. 14). Such behavior was also observed with the ^{60}Co contamination originating from the cooling water of the High-Flux Isotope Reactor (Fig. 19); here, however, dilution at the main channel of White Oak Creek led to a decrease in the ^{60}Co gravel concentrations.

A second type of concentration profile from a point source can be illustrated by the behavior of ^{90}Sr in the Northwest Tributary (Fig. 18). Here a point source of ^{90}Sr entering the creek yielded a

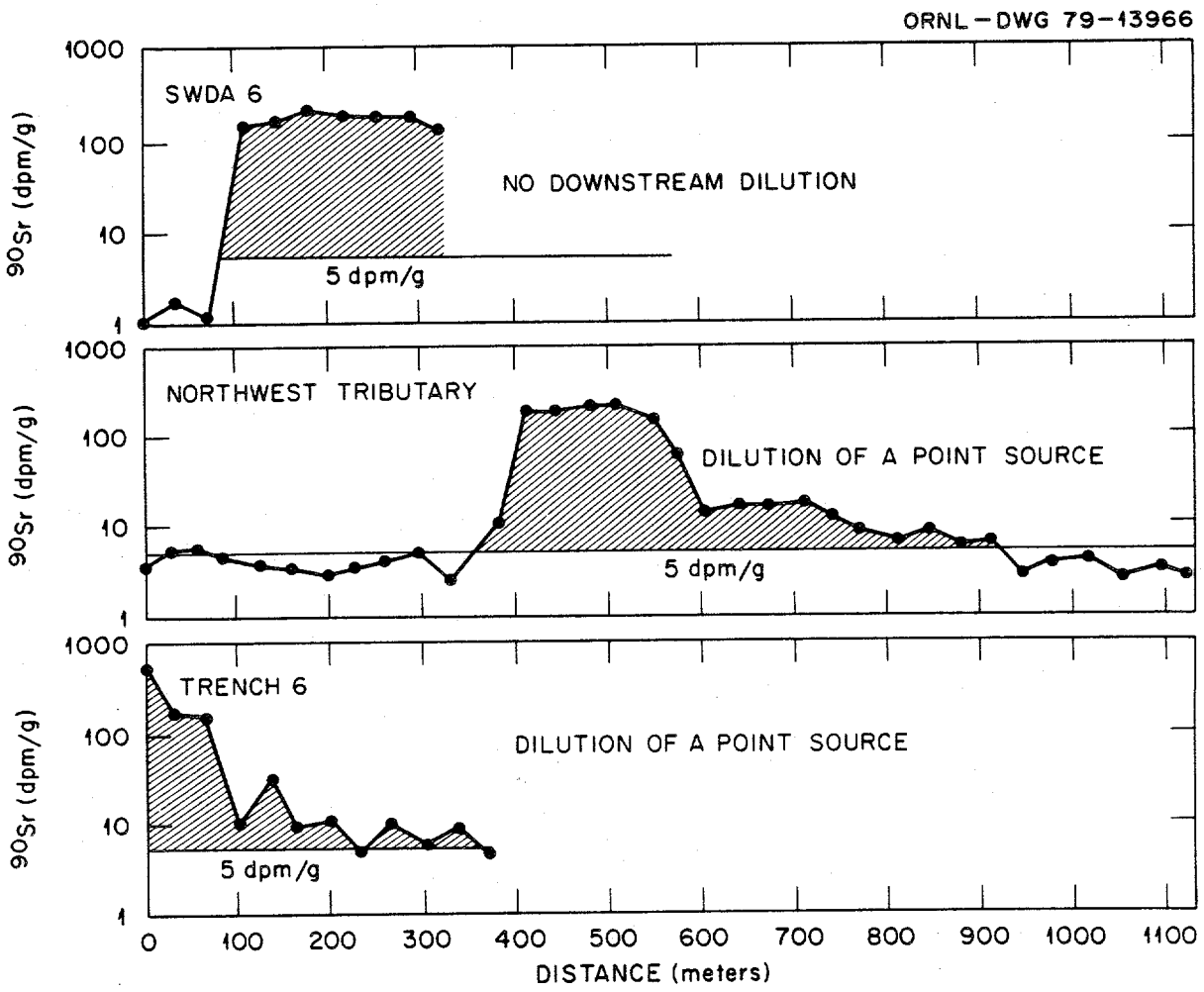

Fig. 18. Types of ^{90}Sr concentration profiles in streambed gravels originating from point sources.

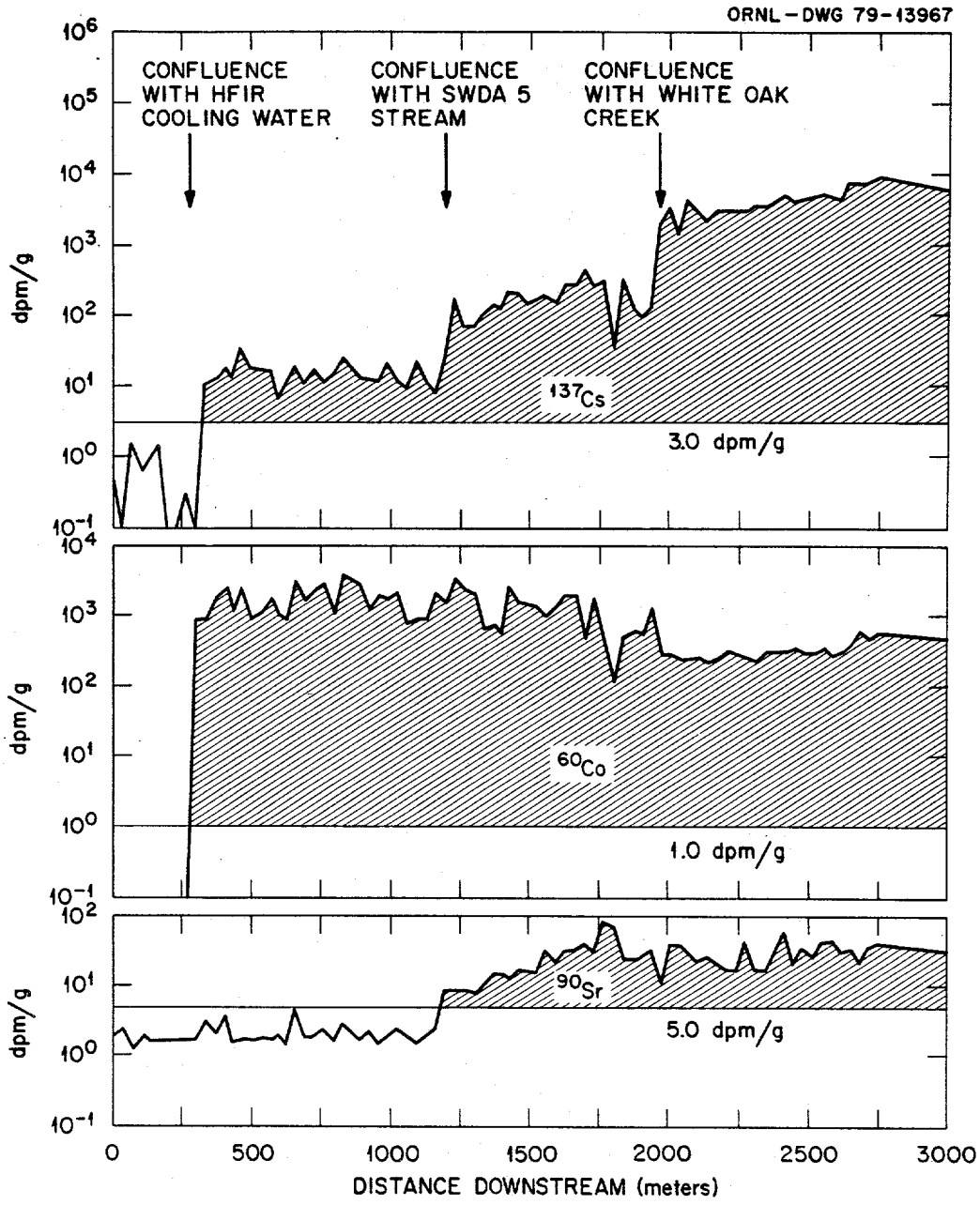

Fig. 19. Behavior of radionuclide concentrations in stream-bed gravels in Melton Branch to inputs from various contamination sources.

constant concentration profile until dilution by the uncontaminated discharge from other tributaries. A similar profile was observed with the point source of ^{90}Sr from trench 6 into a seasonally intermitent stream which joined a larger stream within 100 m (Fig. 18).

Diffuse sources of ^{90}Sr gave rise to concentration profiles which exhibited a gradual downstream increase. An important example of this is illustrated by the reach of Melton Branch on the south side of SWDA 5 between the confluence with the stream draining to the east of SWDA 5 and its confluence with the main channel of White Oak Creek (Fig. 19). A second example of this type of diffuse ^{90}Sr source was observed in the upper half of the stream draining from SWDA 4 (Fig. 10). In the main channel of White Oak Creek below its confluence with Melton Branch (hence, below all major point and diffuse sources), the concentration profile of ^{90}Sr appeared to be quite constant (Fig. 9); the concentrations below White Oak Dam were quite similar to those immediately above White Oak Lake, implying that this settling pond served neither as a sink nor a source of ^{90}Sr.

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APPENDIX

Description of samples analyzed in this study: MONTH, YEAR are the month and year when the sample was taken; NORTH, EAST are the coordinates for the same location on the grid in Figure 20; MN, FE are the Manganese and Iron concentrations of the gravel samples in micrograms per gram of gravel; C060, CS137, SR90 are the ^{60}Co, ^{137}Cs, and ^{90}Sr concentrations of the gravel in disintegrations per minute per gram of gravel.

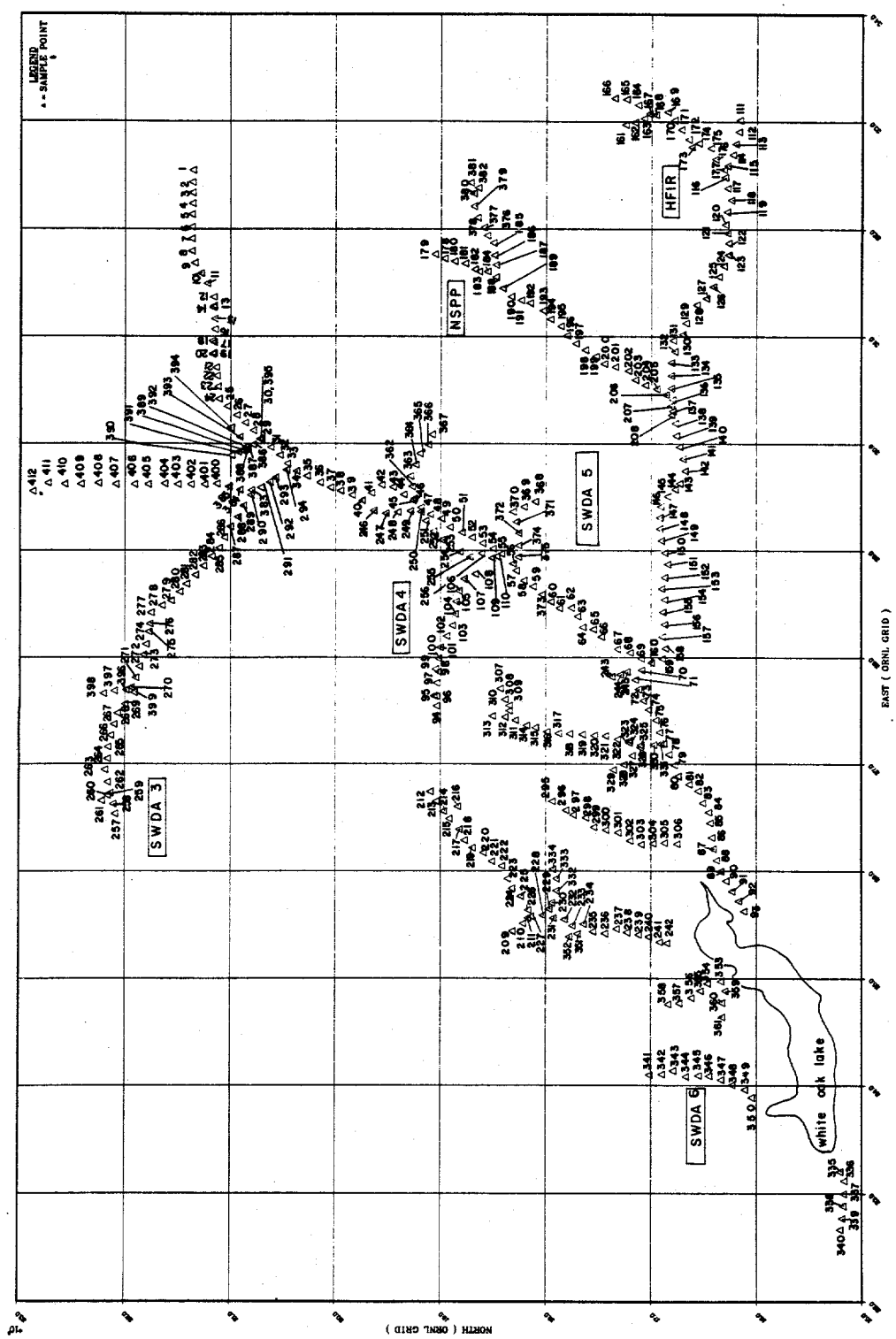

Fig. 20. Sampling locations in White Oak Creek Watershed.

APPENDIX (continued)

SAMPLE	MONTH	YEAR	NORTH	EAST	MN	FE	CO60	CS137	SR90
1	10	78	21360	32545	1220	770	0.0	0.0	0.9
2	10	78	21365	32435	910	800	0.0	0.0	0.2
3	10	78	21365	32335	2040	1010	0.0	0.3	0.1
4	10	78	21365	32235	1910	995	0.0	0.0	0.4
5	10	78	21370	32135	770	665	0.0	1.7	0.1
6	10	78	21370	32020	600	565	0.0	0.0	0.4
7	10	78	21370	31910	750	540	0.0	0.0	0.0
8	10	78	21370	31795	780	635	0.0	0.0	0.5
9	10	78	21345	31685	1130	945	1.3	13.0	3.3
10	10	78	21285	31590	570	560	0.0	8.5	0.5
11	10	78	21225	31495	1350	745	1.6	10.8	1.0
12	10	78	21165	31365	780	460	0.0	6.2	1.2
13	10	78	21160	31285	920	690	1.0	4.4	1.9
14	10	78	21170	31270	422	1120	0.0	15.4	22.4
15	10	78	21155	31165	1300	775	3.1	7.6	2.8
16	10	78	21155	31065	630	720	0.0	52.4	3.2
17	10	78	21160	30945	1030	890	13.3	558.0	5.3
18	10	78	21175	30960	476	440	25.9	72.5	18.2
19	10	78	21165	30845	465	610	17.6	2300.0	4.5
20	10	78	21175	30835	128	355	370.0	1190.0	2.5
21	10	78	21140	30710	595	790	116.0	1690.0	9.7
22	10	78	21135	30630	389	755	127.0	1200.0	9.3
23	10	78	21125	30530	1050	970	147.0	1470.0	6.8
24	10	78	21115	30415	605	895	112.0	3470.0	9.7
25	10	78	21035	30345	695	615	94.0	2370.0	4.5
26	10	78	20945	30260	585	535	98.0	2620.0	3.9
27	10	78	20865	30190	685	790	110.0	1500.0	8.9
28	10	78	20785	30120	420	810	186.0	4720.0	8.4
29	10	78	20705	30045	420	765	164.0	6280.0	6.6
30	10	78	20725	30045	605	1000	130.0	3030.0	18.2
31	10	78	20620	29965	485	1000	93.4	3110.0	6.7
32	10	78	20535	29890	380	1150	195.0	23200.0	17.2
33	10	78	20460	29810	465	1080	190.0	6990.0	12.6
34	10	78	20375	29740	1270	1040	25.8	126.0	5.1
35	10	78	20275	29695	500	1090	118.0	2830.0	8.2
36	10	78	20150	29635	475	1060	117.0	3520.0	7.7
37	10	78	20050	29590	630	950	165.0	6970.0	111.8
38	10	78	19955	29560	760	1290	42.5	1590.0	11.7
39	10	78	19845	29520	585	1080	86.3	638.0	12.0
40	10	78	19750	29470	510	1390	37.7	1090.0	10.7
41	10	78	19660	29540	355	975	94.7	775.0	8.9
42	10	78	19565	29615	365	840	67.1	815.0	7.2
43	10	78	19455	29595	670	1030	105.0	833.0	6.9
44	10	78	19355	29525	845	950	93.9	938.0	7.7
45	10	78	19265	29470	890	1210	45.9	430.0	8.5
46	10	78	19255	29490	1150	1870	0.0	2.8	0.5
47	10	78	19190	29405	1010	890	42.5	356.0	8.9
48	10	78	19100	29340	1090	1280	103.0	1780.0	9.9
49	10	78	18995	29300	845	1240	69.1	661.0	9.0

APPENDIX (continued)

SAMPLE	MONTH	YEAR	NORTH	EAST	MN	FE	CO60	CS137	SR90
50	10	78	18915	29220	790	1330	93.9	748.0	12.1
51	10	78	18800	29170	735	1280	90.4	1060.0	8.5
52	10	78	18705	29125	690	1270	102.0	648.0	7.6
53	10	78	18605	29070	685	1350	57.0	771.0	10.2
54	10	78	18505	29020	965	1400	104.0	1680.0	12.3
55	10	78	18425	28970	895	1440	82.5	1080.0	10.2
56	10	78	18315	28875	740	1310	52.3	635.0	9.2
57	10	78	18285	28810	670	1460	83.3	1110.0	9.4
58	10	78	18210	28720	480	1610	99.4	1710.0	7.2
59	10	78	18125	28665	1040	2140	132.0	1920.0	13.6
60	10	78	17955	28520	810	1570	103.0	1110.0	11.2
61	10	78	17875	28460	1020	1650	106.0	1820.0	19.6
62	10	78	17760	28465	800	1840	118.0	1670.0	8.0
63	10	78	17700	28385	950	1700	144.0	1860.0	12.0
64	10	78	17650	28285	770	1900	166.0	3020.0	8.8
65	10	78	17550	28265	815	1810	200.0	1990.0	18.5
66	10	78	17475	28195	1110	1470	232.0	2450.0	32.1
67	10	78	17320	28080	615	1420	206.0	2520.0	11.5
68	10	78	17200	28045	555	1250	222.0	2510.0	12.5
69	10	78	17095	27990	745	1970	205.0	2400.0	12.9
70	10	78	17090	27880	805	1420	277.0	1910.0	11.6
71	10	78	17150	27790	970	2000	278.0	3270.0	41.0
72	10	78	17120	27700	715	2690	238.0	1460.0	35.7
73	10	78	17075	27595	1060	2500	254.0	4110.0	29.6
74	10	78	17025	27510	895	2080	260.0	2920.0	21.5
75	10	78	16955	27410	1090	2680	218.0	2160.0	25.5
76	10	78	16920	27295	925	2150	254.0	3010.0	20.9
77	10	78	16880	27190	990	2180	315.0	2990.0	16.9
78	10	78	16830	27090	935	2350	266.0	2970.0	16.0
79	10	78	16780	26990	1030	2260	239.0	2850.0	43.1
80	10	78	16745	26885	950	2360	224.0	3580.0	17.6
81	10	78	16640	26810	990	2310	304.0	3290.0	16.5
82	10	78	16555	26750	975	1830	303.0	3820.0	28.2
83	10	78	16510	26640	1160	2420	298.0	4900.0	59.4
84	10	78	16455	26550	1160	2170	340.0	4060.0	19.7
85	10	78	16440	26450	1010	2020	297.0	4020.0	35.2
86	10	78	16425	26315	1040	2390	290.0	4560.0	25.6
87	10	78	16410	26210	1110	3020	344.0	5030.0	43.0
88	10	78	16380	26105	1110	2470	272.0	4560.0	45.8
89	10	78	16340	26000	865	2550	312.0	4070.0	29.4
90	10	78	16290	25910	1010	2790	397.0	7360.0	33.4
91	10	78	16235	25815	820	2590	594.0	7050.0	20.8
92	10	78	16170	25725	870	3390	474.0	7900.0	33.3
93	10	78	16120	25630	775	3920	570.0	8840.0	38.1
94	10	78	19040	27545	215	4370	0.0	18.6	167.0
95	10	78	19065	27640	2710	5570	0.5	87.4	295.7
96	10	78	19045	27635	780	4230	0.0	17.5	386.1
97	10	78	19040	27760	7610	2990	0.0	156.0	583.6
98	10	78	19025	27865	6270	2830	0.0	79.6	687.5

APPENDIX (continued)

SAMPLE	MONTH	YEAR	NORTH	EAST	MN	FE	CO60	CS137	SR90
99	10	78	19050	27890	1270	6090	0.0	140.0	1648.3
100	10	78	19020	27990	4110	2690	0.0	64.4	1018.3
101	10	78	19005	28095	4820	3680	0.0	82.9	1795.1
102	10	78	18945	28200	3080	5190	11.1	274.0	1512.8
103	10	78	18890	28300	9940	4740	21.8	423.0	1637.0
104	10	78	18865	28410	9250	4300	12.6	361.0	1828.9
105	10	78	18855	28520	6130	4860	3.4	112.0	1126.7
106	10	78	18830	28625	9390	5170	6.0	194.0	1693.5
107	10	78	18780	28735	6070	3500	1.7	61.0	1049.9
108	10	78	18675	28780	9830	3370	19.0	113.0	1298.3
109	10	78	18515	28930	295	1660	192.0	4020.0	119.6
110	10	78	18470	28945	350	3250	201.0	3210.0	141.1
111	10	78	16165	33015	2550	2520	0.0	1.5	1.9
112	10	78	16175	32910	2503	2560	0.0	1.7	2.2
113	10	78	16205	32795	2160	2650	0.0	0.0	1.5
114	10	78	16230	32690	2710	2370	0.0	0.9	2.5
115	10	78	16280	32590	2340	2480	0.0	0.0	1.5
116	10	78	16305	32485	1950	2970	1110.0	12.9	1.4
117	10	78	16280	32380	2060	2720	2380.0	33.0	1.6
118	10	78	16245	32270	2400	2350	864.0	18.2	1.5
119	10	78	16285	32160	2240	2460	1100.0	16.4	1.8
120	10	78	16300	32045	2080	2400	1700.0	16.0	1.5
121	10	78	16280	31965	2280	2600	1040.0	6.5	2.0
122	10	78	16270	31870	1620	1840	851.0	10.1	1.3
123	10	78	16270	31760	2400	2930	3120.0	17.4	4.2
124	10	78	16315	31660	2000	2500	1670.0	10.8	1.8
125	10	78	16360	31560	1720	2220	2300.0	16.6	1.9
126	10	78	16415	31465	2140	2580	2900.0	11.4	2.3
127	10	78	16480	31355	1970	2450	1060.0	15.5	1.5
128	10	78	16570	31295	2150	2840	3910.0	25.4	3.0
129	10	78	16675	31125	1640	1820	2780.0	13.2	1.6
130	10	78	16705	31020	1500	1570	1220.0	12.7	2.2
131	10	78	16795	30965	1870	1490	1920.0	11.9	1.4
132	10	78	16780	30860	1770	1850	1710.0	21.2	1.9
133	10	78	16815	30755	2280	2430	2120.0	12.1	2.4
134	10	78	16815	30635	1680	1870	759.0	9.4	1.9
135	10	78	16815	30515	1770	2380	883.0	22.2	1.4
136	10	78	16800	30410	2190	1780	848.0	11.5	1.9
137	10	78	16800	30300	1950	1870	2070.0	7.8	2.3
138	10	78	16775	30185	2530	1900	1520.0	26.3	8.9
139	10	78	16765	30075	2740	2190	3320.0	174.0	9.1
140	10	78	16735	29970	2360	1920	2400.0	67.5	8.6
141	10	78	16700	29855	2190	1830	2050.0	70.0	7.7
142	10	78	16680	29745	2790	2380	622.0	106.0	10.7
143	10	78	16730	29630	2560	2530	750.0	141.0	14.6
144	10	78	16775	29575	2210	2020	560.0	121.0	14.9
145	10	78	16845	29510	2500	2970	2570.0	211.0	12.7
146	10	78	16910	29415	2470	2330	1520.0	192.0	17.1
147	10	78	16920	29310	2480	1880	1440.0	142.0	15.5

APPENDIX (continued)

SAMPLE	MONTH	YEAR	NORTH	EAST	MN	FE	CO60	CS137	SR90
148	10	78	16920	29205	2390	2380	1310.0	170.0	15.6
149	10	78	16905	29095	2460	2000	951.0	189.0	31.6
150	10	78	16870	28980	3590	1640	1310.0	154.0	22.3
151	10	78	16845	28870	3210	2390	2020.0	265.0	32.4
152	10	78	16875	28750	2810	2170	1880.0	257.0	34.2
153	10	78	16900	28645	2320	2280	512.0	411.0	39.9
154	10	78	16875	28540	2450	1990	1760.0	248.0	30.3
155	10	78	16890	28425	2700	2390	512.0	305.0	82.9
156	10	78	16875	28310	2140	2360	121.0	33.0	69.2
157	10	78	16895	28195	1110	1900	540.0	310.0	23.8
158	10	78	16865	28085	1250	1190	610.0	129.0	24.1
159	10	78	16895	27990	1290	1270	554.0	90.7	26.1
160	10	78	17000	27950	1860	1830	1200.0	128.0	32.5
161	10	78	17250	32975	8420	4400	0.0	23.8	2.0
162	10	78	17160	32995	3370	2460	0.0	1.3	1.0
163	10	78	17075	33030	3410	2770	0.0	0.0	1.5
164	10	78	17130	33155	2410	3080	0.0	1.5	1.2
165	10	78	17240	33210	2410	2720	0.0	0.0	2.4
166	10	78	17355	33220	2700	2910	0.0	0.9	1.9
167	10	78	17025	33075	2470	3040	0.0	0.6	1.9
168	10	78	16960	33065	2290	3170	0.0	0.9	1.5
169	10	78	16850	33090	2100	3320	0.0	1.4	1.5
170	10	78	16785	33015	2230	3300	0.0	0.0	1.5
171	10	78	16720	32930	2310	3380	0.0	0.0	1.5
172	10	78	16650	32835	2330	3270	0.0	0.3	1.6
173	10	78	16620	32765	1000	1440	745.0	15.2	1.4
174	10	78	16555	32800	950	3320	881.0	0.0	1.6
175	10	78	16440	32755	1750	3310	921.0	10.5	3.0
176	10	78	16395	32650	1530	1430	1820.0	12.6	2.0
177	10	78	16315	32555	1700	3190	2510.0	18.6	3.8
178	10	78	18970	31725	1370	6990	0.0	0.4	2.0
179	10	78	19060	31765	1170	8160	0.0	1.6	2.0
180	10	78	18875	31700	705	3210	34.8	2.7	3.1
181	10	78	18775	31680	1420	4570	13.3	2.8	3.3
182	10	78	18680	31625	1170	6300	51.6	2.4	10.8
183	10	78	18640	31605	3910	6060	103.0	194.0	14.1
184	10	78	18560	31605	535	6960	12.3	1470.0	42.4
185	10	78	18505	31870	3570	3450	0.0	16.3	52.1
186	10	78	18495	31755	6520	2880	0.0	14.6	73.3
187	10	78	18480	31665	6780	6310	1.9	22.1	70.9
188	10	78	18480	31555	3410	3720	12.0	309.0	62.3
189	10	78	18415	31450	2590	4550	5.5	4860.0	127.5
190	10	78	18335	31370	3840	4250	3.5	7460.0	159.1
191	10	78	18240	31335	3920	3730	2.7	6060.0	159.1
192	10	78	18155	31310	3100	2550	0.0	2620.0	90.8
193	10	78	18030	31240	1630	2610	0.0	3870.0	80.8
194	10	78	17960	31160	2000	2520	2.3	3620.0	121.9
195	10	78	17865	31095	2040	2600	1.6	2880.0	156.9
196	10	78	17805	31010	1790	2460	0.0	1530.0	112.9

APPENDIX (continued)

SAMPLE	MONTH	YEAR	NORTH	EAST	MN	FE	CO60	CS137	SR90
197	10	78	17720	30935	1050	2290	0.0	200.0	87.1
198	10	78	17630	30875	1780	2650	1.1	5580.0	145.6
199	10	78	17530	30815	1790	2570	2.0	2880.0	119.6
200	10	78	17450	30745	1600	3130	0.0	1930.0	79.8
201	10	78	17345	30715	1780	3060	0.0	1700.0	152.4
202	10	78	17220	30680	1130	4290	0.0	1820.0	89.1
203	10	78	17155	30595	1940	3290	0.0	1440.0	115.1
204	10	78	17060	30545	1870	3650	0.0	1610.0	95.9
205	10	78	16950	30510	1840	4140	0.0	1690.0	82.6
206	10	78	16865	30455	2940	3930	0.0	2210.0	104.8
207	10	78	16815	30350	6820	3010	13.0	66.9	126.4
208	10	78	16810	30265	4490	2750	447.0	34.0	62.0
209	10	78	18320	25440	2730	3420	0.0	0.0	2.8
210	10	78	18220	25510	2490	3240	0.0	0.8	2.0
211	10	78	18175	25565	2560	2570	0.0	0.9	3.7
212	10	78	19100	26745	1030	5990	0.0	1.4	1.6
213	10	78	19045	26655	1250	5720	0.0	1.2	7.1
214	10	78	18975	26560	1040	5110	0.0	1.8	27.4
215	10	78	18920	26490	1870	5440	0.0	4.2	83.5
216	10	78	18840	26605	1320	2720	22.1	200.0	846.7
217	10	78	18825	26390	1550	4780	0.0	3.6	43.0
218	10	78	18775	26290	1910	3840	0.0	3.5	20.4
219	10	78	18700	26220	1940	3660	0.0	6.1	61.4
220	10	78	18595	26175	2010	4680	0.0	5.1	118.5
221	10	78	18515	26095	1920	3560	0.0	8.4	50.8
222	10	78	18420	26050	2070	3450	0.0	6.9	57.0
223	10	78	18365	25930	2730	3700	0.0	12.1	42.4
224	10	78	18325	25840	2050	3470	0.0	6.9	43.9
225	10	78	18240	25770	1970	3220	0.0	10.5	47.4
226	10	78	18170	25640	2040	3220	0.0	5.1	58.2
227	10	78	18140	25580	1580	3600	0.0	6.4	43.3
228	10	78	18030	25590	495	2730	0.0	0.3	32.5
229	10	78	17970	25650	1360	2510	41.7	14.6	10.2
230	10	78	17945	25710	1150	2070	23.0	16.9	8.2
231	10	78	17930	25565	555	4440	6.3	12.1	47.1
232	10	78	17825	25550	1870	2450	13.9	3.8	52.8
233	10	78	17750	25490	2110	3170	11.3	10.7	39.1
234	10	78	17645	25505	2410	2490	15.0	8.6	30.5
235	10	78	17555	25435	2510	2690	14.2	9.7	40.5
236	10	78	17440	25420	2370	2440	14.4	6.7	42.2
237	10	78	17330	25460	2040	2320	366.0	4.7	36.9
238	10	78	17230	25430	1880	3230	666.0	15.0	33.0
239	10	78	17120	25410	2180	2680	634.0	0.9	30.0
240	10	78	17025	25390	2060	3740	712.0	1.0	34.6
241	10	78	16920	25340	2330	3260	776.0	8.7	39.9
242	10	78	16850	25330	2520	3370	885.0	14.5	33.9
243	10	78	17375	27820	1660	3470	36700.0	123.0	7.6
244	10	78	17295	27840	905	4260	12900.0	72.2	2.3
245	10	78	17230	27865	1150	4390	16300.0	66.6	4.0

APPENDIX (continued)

SAMPLE	MONTH	YEAR	NORTH	EAST	MN	FE	CO60	CS137	SR90
246	10	78	19640	29375	995	2140	29.2	256.0	15.8
247	10	78	19525	29350	465	1770	110.0	2670.0	10.9
248	10	78	19410	29360	410	1890	87.4	1920.0	19.8
249	10	78	19290	29370	330	1760	165.0	5250.0	25.1
250	10	78	19190	29365	760	1990	477.0	15100.0	32.2
251	10	78	19145	29280	1090	3030	131.0	3240.0	99.0
252	10	78	19070	29180	735	1170	222.0	10400.0	149.0
253	10	78	18990	29100	2320	1740	128.0	3790.0	100.3
254	10	78	18820	28990	420	2700	204.0	5680.0	27.2
255	10	78	18725	28940	400	4260	65.9	970.0	85.3
256	10	78	18615	28975	2740	3350	41.3	409.0	118.5
257	10	78	22095	26535	2960	1470	0.1	2.1	3.7
258	10	78	22110	26630	2660	1290	0.0	2.0	5.3
259	10	78	22140	26730	2160	1020	0.0	0.8	5.8
260	10	78	22165	26710	1060	1380	0.0	0.7	1.3
261	10	78	22220	26660	1210	1030	0.0	0.2	0.1
262	10	78	22165	26830	1550	1220	0.0	1.3	4.0
263	10	78	22190	26945	2150	1450	0.0	0.2	3.4
264	10	78	22165	27050	2160	1480	0.0	0.9	3.2
265	10	78	22160	27160	1270	1020	0.0	0.0	2.9
266	10	78	22135	27265	2060	1410	0.0	1.2	3.6
267	10	78	22105	27370	2200	1510	0.0	1.1	4.1
268	10	78	22070	27475	1910	1100	0.0	0.0	5.0
269	10	78	22000	27560	1340	870	0.0	0.4	2.4
270	10	78	21940	27715	2240	1350	0.0	0.0	10.9
271	10	78	21900	27810	2470	1650	0.0	0.0	187.4
272	10	78	21865	27915	1720	1230	0.5	0.0	189.6
273	10	78	21825	28025	2740	1620	0.0	0.1	213.3
274	10	78	21800	28125	2980	1850	0.0	1.2	214.5
275	10	78	21775	28240	2860	1670	0.0	1.1	146.7
276	10	78	21765	28315	2290	2060	0.0	1.4	72.9
277	10	78	21755	28420	2290	2210	0.0	0.0	13.9
278	10	78	21650	28485	2100	2180	0.0	0.5	17.4
279	10	78	21565	28530	1790	1590	0.0	1.0	16.5
280	10	78	21485	28615	2170	1900	0.0	0.0	18.2
281	10	78	21420	28690	1610	1140	0.0	0.0	13.4
282	10	78	21335	28770	1910	1490	0.0	2.0	8.5
283	10	78	21260	28855	2090	1570	0.0	0.0	6.9
284	10	78	21180	28945	2270	1500	0.0	0.1	9.1
285	10	78	21110	29025	2230	1320	0.6	1.1	6.3
286	10	78	21060	29125	2330	1620	0.0	0.0	7.1
287	10	78	21005	29225	2400	1670	0.0	0.3	2.9
288	10	78	20935	29315	1790	1300	0.0	0.0	4.0
289	10	78	20880	29420	1960	1240	0.0	0.9	4.5
290	10	78	20810	29520	1650	1400	0.0	0.5	2.8
291	10	78	20715	29590	1460	1320	0.0	1.6	3.6
292	10	78	20630	29640	1980	1720	0.0	9.8	2.9
293	10	78	20575	29680	1390	1250	0.0	33.1	3.4
294	10	78	20470	29750	1460	1160	0.0	28.0	3.1

APPENDIX (continued)

SAMPLE	MONTH	YEAR	NORTH	EAST	MN	FE	CO60	CS137	SR90
295	10	78	17940	26655	8060	4140	8.3	10.0	2.2
296	10	78	17810	26570	2740	2200	172.0	28.2	3.1
297	10	78	17740	26525	1510	2100	121.0	4.1	8.2
298	10	78	17620	26500	1670	2310	207.0	9.7	5.1
299	10	78	17545	26410	2280	2530	343.0	14.7	5.0
300	10	78	17430	26385	2620	3260	430.0	13.9	1.6
301	10	78	17315	26360	2480	2120	1380.0	22.8	2.5
302	10	78	17200	26285	210	180	89.5	0.2	0.0
303	10	78	17095	26250	565	660	710.0	16.8	0.6
304	10	78	16980	26260	1330	1770	2070.0	77.0	2.2
305	10	78	16875	26265	1880	4220	2040.0	29.5	2.7
306	10	78	16755	26255	1890	3760	3990.0	70.5	3.2
307	11	78	18435	27705	1840	2320	809.0	1500.0	519.3
308	11	78	18390	27605	2050	2620	55.4	108.0	154.6
309	11	78	18350	27550	620	2860	0.2	10.4	2.7
310	11	78	18350	27500	720	2060	12.0	79.4	143.3
311	11	78	18290	27410	3600	2860	0.0	9.8	9.2
312	11	78	18390	27440	2910	2210	0.0	3.7	1.1
313	11	78	18510	27450	970	1210	0.0	3.5	1.2
314	11	78	18185	27360	2160	2660	1.9	9.8	31.3
315	11	78	18100	27340	2050	2780	0.0	12.8	8.4
316	11	78	17990	27300	3920	3430	8.9	20.6	10.6
317	11	78	17880	27285	1970	3410	0.1	6.0	4.2
318	11	78	17775	27280	2660	3510	0.0	11.5	9.8
319	11	78	17655	27275	2170	3060	1.5	12.3	5.4
320	11	78	17540	27265	1900	3420	6.0	10.5	8.8
321	11	78	17435	27260	1660	4060	148.0	12.2	4.2
322	11	78	17320	27230	1190	3860	294.0	75.4	4.8
323	11	78	17210	27200	1000	3120	198.0	30.0	19.4
324	11	78	17205	27220	1220	3410	308.0	19.7	5.8
325	11	78	17100	27175	1200	3340	216.0	25.4	3.6
326	11	78	17105	27155	1790	2840	328.0	0.0	1.8
327	11	78	17180	27080	1570	2850	210.0	3.3	1.4
328	11	78	17260	26995	1540	2670	158.0	0.1	1.0
329	11	78	17360	26945	915	1430	0.0	7.3	2.0
330	11	78	16975	27180	1280	3270	190.0	21.4	4.9
331	11	78	16910	27190	1700	3150	192.0	33.3	4.4
332	11	78	17900	25820	1110	2720	39.5	21.0	7.3
333	11	78	17900	25930	1860	3030	151.0	42.3	13.8
334	11	78	17935	26020	1230	2800	59.2	34.7	10.4
335	11	78	15210	23205	840	2270	261.0	2150.0	16.2
336	11	78	15170	23120	870	2860	508.0	11800.0	41.3
337	11	78	15175	23000	860	3190	536.0	11500.0	38.1
338	11	78	15180	22890	900	3840	458.0	11900.0	22.3
339	11	78	15190	22775	1000	4830	496.0	9540.0	32.5
340	11	78	15210	22670	1340	4040	258.0	5260.0	36.1
341	11	78	17020	24100	1090	4320	0.0	0.2	1.1
342	11	78	16900	24105	2420	2210	0.0	0.0	1.6
343	11	78	16795	24140	3240	1200	0.0	0.7	1.1

APPENDIX (continued)

SAMPLE	MONTH	YEAR	NORTH	EAST	MN	FE	CO60	CS137	SR90
344	11	78	16675	24085	2040	4360	0.0	0.8	146.7
345	11	78	16555	24095	1660	890	0.0	0.0	167.0
346	11	78	16455	24090	1710	1360	0.0	2.3	221.2
347	11	78	16340	24055	4030	1170	0.0	2.0	188.5
348	11	78	16235	24005	3220	2540	0.0	0.3	176.1
349	11	78	16115	23960	2910	1530	0.0	1.4	179.5
350	11	78	16050	23890	2340	2250	0.0	2.3	130.9
351	11	78	17700	25415	3110	880	0.0	10.9	3.9
352	11	78	17780	25390	2590	655	0.0	4.1	3.3
353	11	78	16340	24975	2760	1400	0.0	1.3	1.4
354	11	78	16465	24955	3470	2310	0.0	0.4	1.1
355	11	78	16535	24885	955	1740	0.0	0.3	0.6
356	11	78	16620	24820	1140	3590	0.0	0.6	1.6
357	11	78	16735	24770	2600	1450	0.0	1.5	1.6
358	11	78	16840	24760	435	1490	0.0	1.5	1.3
359	11	78	16300	24885	1060	665	0.0	2.3	3.0
360	11	78	16330	24775	1900	1050	0.0	1.2	1.6
361	11	78	16330	24640	1030	465	0.0	1.2	1.6
362	2	79	19275	29600	2730	2460	0.0	0.0	1.5
363	2	79	19290	29695	3870	2710	0.0	0.8	1.4
364	2	79	19240	29800	3560	2700	0.0	1.0	2.4
365	2	79	19200	29905	4350	3010	0.0	1.5	1.5
366	2	79	19120	29990	4430	3260	0.0	1.4	1.2
367	2	79	19085	30085	3500	3830	0.0	0.6	1.5
368	2	79	18105	29460	8500	4990	0.0	3.1	27.5
369	2	79	18210	29410	4500	4940	0.0	5.0	19.4
370	2	79	18315	29365	3690	1150	0.0	2.2	8.9
371	2	79	18285	29270	3890	5730	0.0	2.5	9.0
372	2	79	18260	29165	2420	3540	0.0	1.4	7.5
373	10	78	18040	28590	785	1910	90.3	1320.0	18.5
374	2	79	18260	29050	4070	5530	0.0	6.9	8.1
375	2	79	18265	28940	2790	3730	0.0	6.4	9.3
376	2	79	18565	31940	2200	4440	0.0	19.2	70.9
377	2	79	18595	32015	8110	5350	3.6	55.8	58.3
378	2	79	18655	32105	5540	6330	4.7	82.4	89.1
379	2	79	18690	32210	1590	8360	14.0	64.3	22.5
380	2	79	18695	32330	5550	4120	1.5	49.7	75.0
381	2	79	18720	32430	3000	3120	4.8	85.2	166.5
382	2	79	18650	32380	9140	2780	0.0	1.2	2.5
383	2	79	20805	29565	1030	1350	0.0	132.0	2.4
384	2	79	20915	29570	795	1020	0.0	86.7	1.9
385	2	79	21020	29600	675	1270	0.0	272.0	5.5
386	2	79	20790	29990	1540	1720	0.0	33.0	14.2
387	2	79	20840	29955	420	1980	0.0	16.2	10.2
388	2	79	20860	29945	1270	1200	0.0	10.0	15.6
389	2	79	20895	29935	1430	1370	0.0	2.0	1.4
390	2	79	20995	29890	295	2900	0.0	2.9	0.7
391	2	79	20930	29920	1040	1130	0.0	0.4	0.3
392	2	79	20860	29970	1860	1050	0.0	1.5	0.7

APPENDIX (continued)

SAMPLE	MONTH	YEAR	NORTH	EAST	MN	FE	CO60	CS137	SR90
393	2	79	20915	30060	1720	955	0.0	0.0	0.5
394	2	79	20995	30150	4910	2660	0.0	0.8	0.9
395	2	79	20720	30040	1890	2700	29.1	1040.0	30.0
396	2	79	21990	27695	1270	1300	0.0	0.4	0.7
397	2	79	22100	27685	1420	1080	0.0	0.0	2.5
398	2	79	22205	27660	2190	1450	0.0	0.7	1.0
399	2	79	21915	27670	1680	2510	0.0	1.4	3.4
400	2	79	21135	29625	775	1610	0.0	8.4	1.5
401	2	79	21250	29620	545	1310	0.0	5.0	2.2
402	2	79	21380	29620	805	815	0.0	0.0	1.8
403	2	79	21505	29620	670	1110	0.0	0.7	1.1
404	2	79	21630	29625	860	890	0.0	0.4	1.6
405	2	79	21785	29620	430	670	0.0	1.3	1.1
406	2	79	21915	29620	1370	1160	0.0	0.0	2.0
407	2	79	22090	29620	600	1450	0.0	0.4	0.0
408	2	79	22255	29630	540	690	0.0	1.4	1.0
409	2	79	22420	29620	640	1410	0.0	0.9	1.6
410	2	79	22575	29620	300	1100	0.0	0.2	1.2
411	2	79	22735	29630	1380	1580	0.0	1.1	1.3
412	2	79	22880	29555	1060	910	0.0	0.9	1.2

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